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Claims for the following Contracting State: ES.

(54) **N-acyloxy hindered amine stabilizers.**

(57) Hindered amines based on various 2,2,6,6-tetra-alkylated nitrogen-containing heterocyclic moieties wherein the hindered nitrogen atom on the ring is substituted with O-acyl, O-carbamoyl or O-carbonate substituents and the 4-position of the ring is substituted with a variety of groups, are effective as light stabilizers in diverse substrate systems. Some of them are new compounds.

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Description

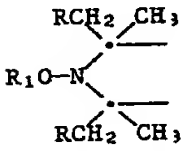
N-ACYLOXY HINDERED AMINE STABILIZERS

The instant invention relates to the stabilization of a wide variety of ambient curable and of acid catalyzed thermosetting coating compositions by the incorporation therein of N-acyloxy-substituted hindered amine light stabilizers as well as to new N-acyloxy hindered amines.

Hindered amine light stabilizers are well known to be effective in stabilizing a host of organic substrates including polymers against the deleterious effects of oxygen and light. Such hindered amine light stabilizers have been used in the stabilization of hot-crosslinkable alkyd or acrylic metallic stoving lacquers (US 4,426,472) and in stabilizing acid-catalyzed stoving lacquers based on hot-crosslinkable acrylic polyester or alkyd resins (US 4,344,876 and 4,426,471). The hindered amine light stabilizers of these patents do not possess structures having an acyloxy group substituted directly on the hindered N-atom of the compound.

Related hindered amine stabilizers have been utilized individually and in combination with ultra-violet light absorbers to improve the performance characteristics of coating systems. Notwithstanding such improvements, there still exists a need to further retard the photooxidation and photodegradation of such coating systems and thereby provide increased effectiveness by maintaining the physical integrity of the coatings. Such effectiveness can be manifested by prevention of embrittlement, cracking, corrosion, erosion, loss of gloss, chalking and yellowing of the coating.

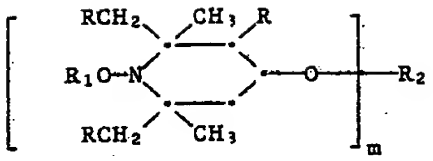
It has now been determined that the aforementioned improvements can be achieved by substitution on the hindered N-atom of the hindered amines with acyloxy groups and the utilization of such derivatives in ambient curable and acid catalyzed thermosetting coating systems. In particular, the physical integrity of the coatings is maintained to a higher degree with significant reduction in loss of gloss and yellowing. Accordingly, the present invention relates to a stabilized ambient curable or acid catalyzed thermosetting coating composition containing an effective stabilizing amount of a hindered amine compound containing the group



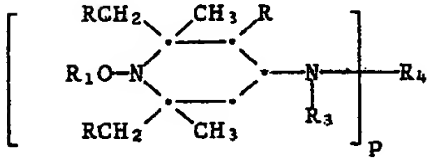
wherein R is hydrogen or methyl and R_1 is a group

, wherein D is C_1-C_{18} alkyl, C_1-C_{18} alkoxy, phenyl, phenyl subtituted by hydroxy, alkyl or alkoxy, or D is amino or amino mono- or disubstituted by alkyl or phenyl.

More particularly, the instant invention relates to the use of a compound having one of formulae A to N



(A)



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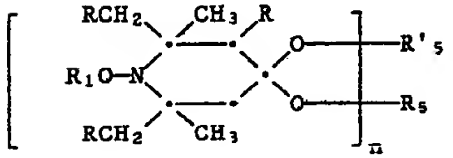

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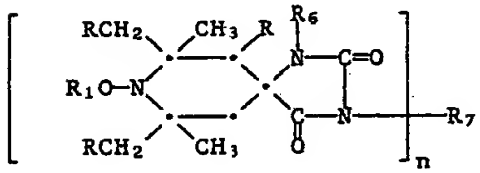

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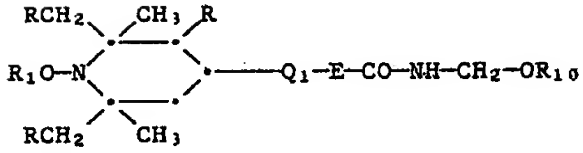

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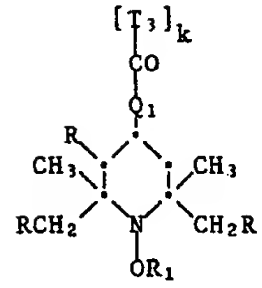

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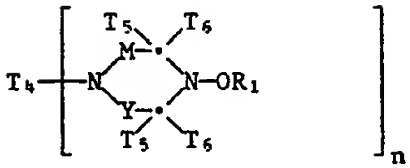

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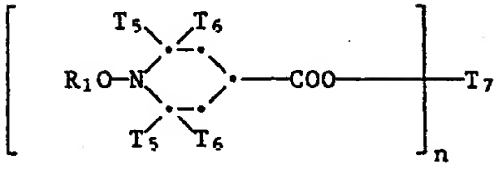

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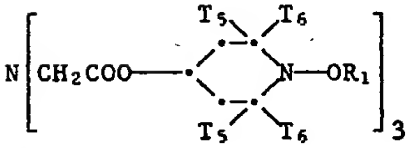

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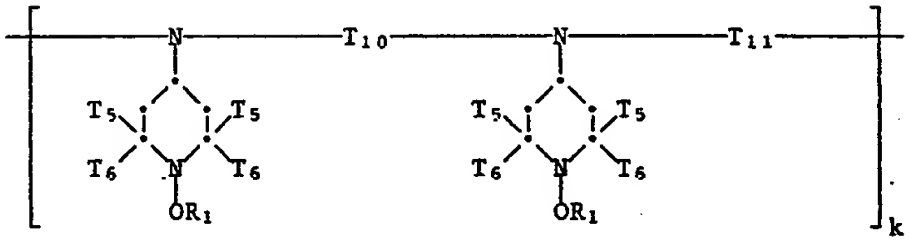

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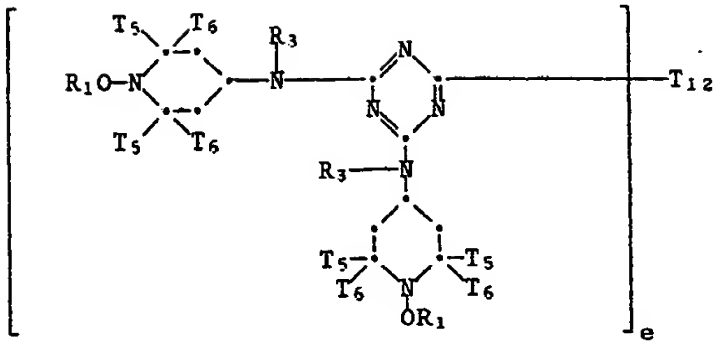

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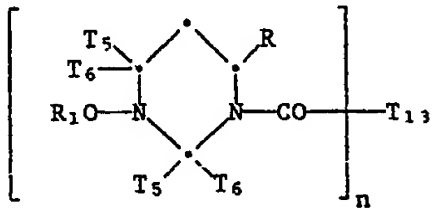

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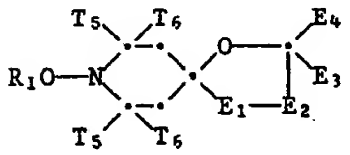

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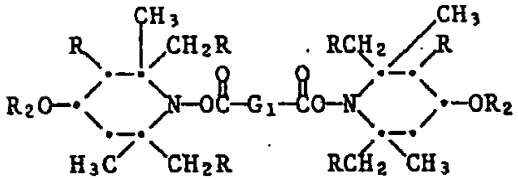
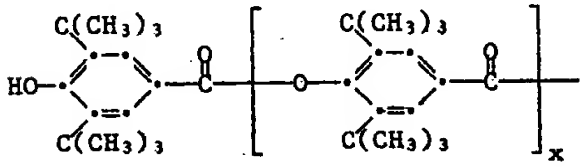
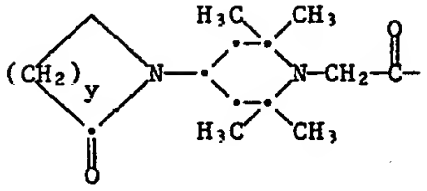


(N)

wherein
R is hydrogen or methyl,
R_1 is a group D-CO- , wherein D is C_1-C_{18} alkyl, C_1-C_{18} alkoxy, phenyl, phenyl substituted by hydroxy, C_1-C_4 alkyl or C_1-C_4 alkoxy, or amino or amino mono- or disubstituted by C_1-C_{12} alkyl or phenyl;
m is 1-4,
when m is 1,
R_2 is hydrogen, C_1-C_{18} alkyl optionally interrupted by one or more oxygen atoms, C_2-C_{12} alkenyl, C_6-C_{10} aryl, C_7-C_{18} aralkyl, glycidyl, a monovalent acyl radical of an aliphatic, cycloaliphatic, araliphatic or aromatic carboxylic acid, or of a carbamic acid, preferably an acyl radical of an aliphatic carboxylic acid having 2-18 C atoms, of a cycloaliphatic carboxylic acid having 6-12 C atoms or of an aromatic carboxylic acid having 7-15 C atoms, or R_2 is a group

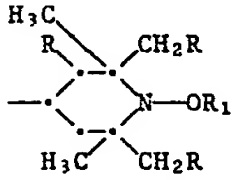


wherein x is 0 or 1, or is a group

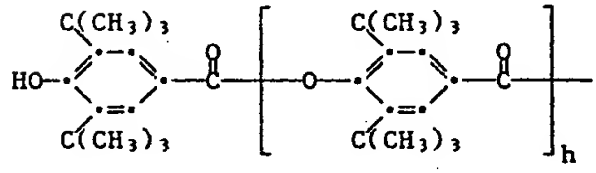


wherein y is 2-4;
when m is 2,
R_2 is C_1-C_{12} alkylene, C_4-C_{12} alkenylene, xylylene, a divalent acyl radical of an aliphatic, cycloaliphatic, araliphatic or aromatic dicarboxylic acid or of a dicarbamic acid, preferably an acyl radical of an aliphatic dicarboxylic acid having having 2-18 C atoms, of an cycloaliphatic or aromatic dicarboxylic acid having 8-14 C atoms, or of an aliphatic, cycloaliphtic or aromatic dicarbamic acid having 8-14 C atoms; or R_2 is a group

wherein D_1 and D_2 independently are hydrogen, an alkyl radical containing up to 8 carbon atoms, phenyl, benzyl or 3,5-di-t-butyl-4-hydroxybenzyl, D_3 is an alkyl or alkenyl radical containing up to 18 carbon atoms; when m is 3, R_2 is a trivalent acyl radical of an aliphatic, cycloaliphatic, or aromatic tricarboxylic acid; when m is 4, R_2 is a tetravalent acyl radical of a saturated or unsaturated aliphatic or aromatic tetracarboxylic acid including 1,2,3,4-butanetetracarboxylic acid, 1,2,3,4-but-2-enetetracarboxylic acid, and 1,2,3,5- and 1,2,4,5-pentanetetracarboxylic acid;
p is 1, 2 or 3,
R_3 is hydrogen, C_1-C_{12} alkyl, C_5-C_7 cycloalkyl, C_7-C_9 aralkyl, C_2-C_{18} alkanoyl, C_3-C_5 alkenoyl or benzoyl; when p is 1,
R_4 is hydrogen, C_1-C_{18} alkyl, C_5-C_7 cycloalkyl, C_2-C_8 alkenyl unsubstituted or substituted by a cyano, carbonyl or carbamide group, or it is aryl, aralkyl, glycidyl, a group of the formula $-CH_2-CH(OH)-Z$ or $-CONH-Z$ wherein Z is hydrogen, methyl or phenyl;
or R_4 is a group of the formula I



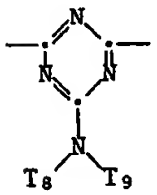
(I) or



with h as 0 or 1;

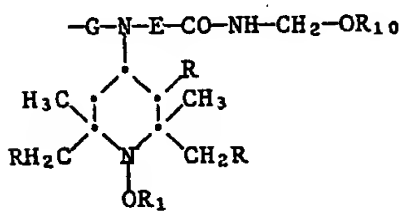
or R_3 and R_4 together are alkylene of 4 to 6 carbon atoms or 1-oxo alkylene or the divalent acyl radical of an aliphatic or aromatic 1,2- or 1,3-dicarboxylic acid;
when p is 2,
R_4 is C_1-C_{12} alkylene, C_6-C_{12} arylene, xylylene, a $-CH_2CH(OH)-CH_2$ group, or a group $-CH_2-CH(OH)-CH_2-$

O-X-O-CH_2-CH(OH)-CH_2- wherein X is C_2-C_{10} alkylene, C_6-C_{15} arylene or C_6-C_{12} cycloalkylene; or, provided that R_3 is not alkanoyl, alkenoyl or benzoyl, R_4 can also be a divalent acyl radical of an aliphatic, cycloaliphatic or aromatic dicarboxylic or dicarbamic acid, or can be the group -CO-, or R_4 is a group of formula II



(II)

where T_8 and T_9 are independently hydrogen, alkyl of 1 to 18 carbon atoms, or T_8 and T_9 together are alkylene of 4 to 6 carbon atoms or 3-oxapentamethylene, preferably T_8 and T_9 together are 3-oxapentamethylene;
when p is 3, R_4 is 2,4,6-triazinetriyl;
n is 1 or 2 and
when n is 1,
R_5 and R'_5 are independently C_1-C_{12} alkyl, C_2-C_{12} alkenyl, C_7-C_{12} aralkyl, or R_5 is also hydrogen, or R_5 and R'_5 together are C_2-C_8 alkylene or hydroxyalkylene or C_4-C_{22} acyloxyalkylene;
when n is 2,
R_5 and R'_5 together are $(-CH_2)_2C(CH_2-)_2$;
R_6 is hydrogen, C_1-C_{12} alkyl, allyl, benzyl, glycidyl or C_2-C_6 alkoxyalkyl;
when n is 1,
R_7 is hydrogen, C_1-C_{12} alkyl, C_3-C_5 alkenyl, C_7-C_9 aralkyl, C_5-C_7 cycloalkyl, C_2-C_4 hydroxyalkyl, C_2-C_6 alkoxyalkyl, C_6-C_{10} aryl, glycidyl, a group of the formula $-(CH_2)_t$-COO-Q or of the formula $-(CH_2)_t$-O-CO-Q wherein t is 1 or 2, and Q is C_1-C_4 alkyl or phenyl; or
when n is 2,
R_7 is C_2-C_{12} alkylene, C_6-C_{12} arylene, a group $-CH_2CH(OH)-CH_2-O-X-O-CH_2-CH(OH)-CH_2-$ wherein X is C_2-C_{10} alkylene, C_6-C_{15} arylene or C_6-C_{12} cycloalkylene, or a group $-CH_2CH(OZ')CH_2-(OCH_2-CH(OZ')CH_2)_2-$ wherein Z' is hydrogen, C_1-C_{18} alkyl, allyl, benzyl, C_2-C_{12} alkanoyl or benzoyl;
Q_1 is -N(R_8)- or -O-;
E is C_1-C_3 alkylene, the group $-CH_2-CH(R_9)-O-$ wherein R_9 is hydrogen, methyl or phenyl, or E is the group $-(CH_2)_3-NH-$ or a direct bond;
R_{10} is hydrogen or C_1-C_{18} alkyl;
R_8 is hydrogen, C_1-C_{18} alkyl, C_5-C_7 cycloalkyl, C_7-C_{12} aralkyl, cyanoethyl, C_6-C_{10} aryl, the group $-CH_2-CH(R_9)-OH$ wherein R_9 has the meaning defined above; a group of the formula I
or a group of the formula



wherein G is C_2-C_8 alkylene or C_6-C_{12} arylene; or R_8 is a group -E-CO-NH-CH_2-OR_{10};

Formula F denotes a recurring structural unit of a polymer where T_3 is ethylene or 1,2-propylene, or is the repeating structural unit derived from an alpha-olefin copolymer with an alkyl acrylate or methacrylate; preferably a copolymer of ethylene and ethyl acrylate, and where k is 2 to 100;
T_4 has the same meaning as R_4 when p is 1 or 2,
T_5 is methyl,
T_6 is methyl or ethyl, or T_5 and T_6 together are tetramethylene or pentamethylene, preferably T_5 and T_6 are each methyl,
M and Y are independently methylene or carbonyl, preferably M is methylene and Y is carbonyl, and T_4 is ethylene where n is 2;
T_7 is the same as R_7, and T_7 preferably octamethylene where n is 2;
T_{10} and T_{11} are independently alkylene of 2 to 12 carbon atoms, or T_{11} is a group of formula II;
e is 2, 3 or 4 and

T_{12} is a group $-N(R_5)-(CH_2)_d-N(R_5)-$ or

$-NH(CH_2)_a-\overset{|}{N}(CH_2)_b-\overset{|}{N}[(CH_2)_c-\overset{|}{N}]_fH$ where a, b and c are independently 2 or 3, d is 2 to 10 and f is 0 or 1, preferably a and c are each 3, b is 2 and f is 1;

T_{13} is the same as R_4 with the proviso that T_{13} cannot be hydrogen when n is 1;

E_1 and E_2, being different, each are -CO- or $-N(E_5)-$, where E_5 is hydrogen, C_1-C_{12} alkyl or alkoxycarbonylalkyl or 4 to 22 carbon atoms, preferably E_1 is -CO- and E_2 is $-N(E_5)-$;

E_3 is hydrogen, alkyl of 1 to 30 carbon atoms, phenyl, naphthyl, said phenyl or said naphthyl substituted by chlorine or by alkyl of 1 to 4 carbon atoms, or phenylalkyl of 7 to 12 carbon atoms, or said phenylalkyl substituted by alkyl of 1 to 4 carbon atoms,

E_4 is hydrogen, alkyl of 1 to 30 carbon atoms, phenyl, naphthyl or phenylalkyl of 7 to 12 carbon atoms, or E_3 and E_4 together are polymethylene of 4 to 17 carbon atoms, or said polymethylene substituted by up to four alkyl groups of 1 to 4 carbon atoms, preferably methyl, and

R_2 of formula (N) is as previously defined when m is 1, G_1 is a direct bond, C_1-C_{12} alkylene, phenylene or -NH-G'-NH wherein G' is C_1-C_{12} alkylene.

In the structures A to N, if any substituents are C_1-C_{18} alkyl, they are for example methyl, ethyl, n-propyl, n-butyl, sec-butyl, tert-butyl, n-hexyl, n-octyl, 2-ethylhexyl, n-nonyl, n-decyl, n-undecyl, n-dodecyl, n-tridecyl, n-tetradecyl, n-hexadecyl or n-octadecyl.

If R_2 is a monovalent acyl radical of a carboxylic acid, it is for example an acyl radical of acetic acid, stearic acid, salicylic acid, methacrylic acid, benzoic acid or β-(3,5-di-tert-butyl-4-hydroxyphenyl)-propionic acid.

If R_2 is a divalent acyl radical of a dicarboxylic acid, it is for example an acyl radical of adipic acid, succinic acid, suberic acid sebacic acid, phthalic acid, maleic acid, dibutylmalonic acid, dibenzylmalonic acid or butyl-(3,5-di-tert-butyl-4-hydroxybenzyl)-malonic acid, or bicycloheptenedicarboxylic acid.

If R_2 is a divalent acyl radical of a dicarbamic acid, it is for example an acyl radical of hexamethylenedicarbamic acid or of 2,4-toluylenedicarbamic acid.

As C_7-C_9 aralkyl, R_3 is particularly phenetyl or above all benzyl.

As C_2-C_{18} alkanoyl, R_3 is for example propionyl, butyryl, octanoyl, dodecanoyl, hexadecanoyl, octadecanoyl, but preferably acetyl; and a C_3-C_5 alkenoyl, R_3 is in particular acryloyl.

If R_4 is C_2-C_8 alkenyl unsubstituted or substituted by a cyano, carbonyl or carbamide group, it is for example 1-propenyl, allyl, methallyl, 2-butenyl, 2-pentenyl, 2-hexenyl, 2-octenyl, 2,2-dicyanovinyl, 1-methyl-2-cyano-2-methoxycarbonyl-vinyl or 2,2-diacetylaminovinyl.

If any substituents are C_2-C_{12} alkylene, they are for example ethylene, propylene, 2,2-dimethylpropylene, tetramethylene, hexamethylene, octamethylene, decamethylene or dodecamethylene.

If any substituents are C_6-C_{15} arylene, they are for example o-, m- or p-phenylene, 1,4-naphthylene or 4,4'-diphenylene.

As C_6-C_{12} cycloalkylene, X is especially cyclohexylene.

If R_5 is C_2-C_8 alkylene or hydroxyalkylene, it is for example ethylene, 1-methyl-ethylene, propylene, 2-ethylpropylene or 2-ethyl-2-hydroxymethylpropylene.

As C_4-C_{22} acyloxyalkylene, R_5 is for example 2-ethyl-2-acetoxymethylpropylene.

If any substituents are C_2-C_8 alkoxyalkyl, they are for example methoxymethyl, ethoxymethyl, propoxymethyl, tert-butoxyethyl, ethoxyethyl, ethoxypropyl, n-butoxyethyl, tert-butoxyethyl, isopropoxyethyl or propoxypropyl.

If R_7 is C_3-C_5 alkenyl, it is for example 1-propenyl, allyl, methallyl, 2-butenyl or 2-pentenyl.

As C_7-C_9 aralkyl, R_7 is in particular phenethyl or above all benzyl; and as C_5-C_7 cycloalkyl, R_7 is especially cyclohexyl.

If R_7 is C_2-C_4 hydroxyalkyl, it is for example 2-hydroxyethyl, 2-hydroxypropyl, 3-hydroxypropyl, 2-hydroxybutyl or 4-hydroxybutyl.

As C_6-C_{10} aryl, R_7 is in particular phenyl, or alpha- or β-naphthyl which is unsubstituted or substituted by halogen or C_1-C_4 alkyl.

If R_7 is C_2-C_{12} alkylene, it is for example ethylene, propylene 2,2-dimethylpropylene, tetramethylene, hexamethylene, octamethylene, decamethylene or dodecamethylene.

If R_7 is C_6-C_{12} arylene, it is for example o-, m- or p-phenylene, 1,4-naphthylene or 4,4'-diphenylene.

If Z' is C_2-C_{12} alkanoyl, it is for example propionyl, butyryl, octanoyl, dodecanoyl or preferably acetyl.

As C_5-C_7 cycloalkyl, R_8 is in particular cyclohexyl.

As C_6-C_{10} aryl, R_8 is particularly phenyl, or alpha- or β-naphthyl which is unsubstituted or substituted with halogen or C_1-C_4 alkyl.

As C_1-C_3 alkylene, E is for example methylene, ethylene or propylene.

As C_2-C_8 alkylene, G is for example ethylene, propylene, 2,2-dimethylpropylene, tetramethylene or hexamethylene; and as C_6-C_{12} arylene, G is o-, m- or p-phenylene, 1,4-naphthylene or 4,4'-diphenylene.

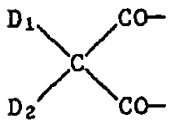
Preferred hindered amine compounds are the compounds of formulae A, B, D, J, K or M wherein R is hydrogen and T_5 and T_6 are methyl.

Further preferred are the compounds of formula A wherein R is hydrogen,

R_1 is a group D-CO- and D is C_1-C_{12} alkyl, C_1-C_4 alkoxy, phenyl, amino or amino monosubstituted by C_1-C_{12} alkyl or phenyl,

m is 1, 2 or 4 and

when m is 1,
R_2 is an acyl radical of an aliphatic C_2-C_{18} carboxylic acid, of a cycloaliphatic C_6-C_{12} carboxylic acid or of an aromatic C_7-C_{15} carboxylic acid and when m is 2, R_2 is a divalent acyl radical of an aliphatic C_2-C_{18} dicarboxylic acid or of a cycloaliphatic or aromatic C_8-C_{14} dicarboxylic acid, or of an aliphatic, cycloaliphatic or aromatic C_8-C_{14} dicarbamic acid, or R_2 is a group



wherein D_1 is C_1-C_8 alkyl or 3,5-di-tert.-butyl-4-hydroxybenzyl und D_2 is D_1 or hydrogen and when m is 4, R_2 is a tetravalent acyl radical of a butane- or pentane-tetracarboxylic acid.

Further preferred are the compounds of formula B wherein R is hydrogen,
R_1 is a group D-CO- and D is C_1-C_{12} alkyl, C_1-C_4 alkoxy, phenyl, amino or amino monosubstituted by C_1-C_{12} alkyl or phenyl,
p is 1 or 2, R_3 is hydrogen, C_1-C_{12} alkyl or C_2-C_{12} alkanoyl and when p is 1, R_4 is C_1-C_{12} alkyl, C_5-C_7 cycloalkyl or a group of formula I, and when p is 2, R_4 is C_2-C_8 alkylene, phenylene or xylylene, and if R_3 is not alkanoyl, R_4 may also be a divalent acyl residue of an aliphatic C_2-C_{10} dicarboxylic acid or of an aromatic C_6 dicarboxylic acid or of an aliphatic or aromatic C_8-C_{15} dicarbamic acid.

The following compounds are examples of hindered amine derivatives applicable for use in the invention:

1. di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) phthalate
2. di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) isophthalate
3. di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) sebacate
4. di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) succinate
5. di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) adipate
6. di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) n-butylmalonate
7. di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) 2,2-diethylmalonate
8. poly-{[6-[(1,1,3,3-tetramethylbutyl)-imino]-1,3,5-triazine-2,4-diyl]-[2-(1-acetoxy-2,2,6,6-tetramethylpiperidyl)-imino]-hexamethylene-[4-(1-acetoxy-2,2,6,6-tetramethylpiperidyl)-imino]}
9. 1,4-diacetoxy-2,2,6,6-tetramethylpiperidine
10. 1,4-acetoxy-4-hydroxy-2,2,6,6-tetramethylpiperidine
11. 1,4-acetoxy-4-acryloxy-2,2,6,6-tetramethylpiperidine
12. polymer of 11
13. (1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl)4-hydroxy-3,5-di-tert.butylbenzoate
14. di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) 2-(4-hydroxy-3,5-di-tert.butylbenzyl)-n-butylmalonate
15. N-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl)-N-(n-butyl)-4-(4-hydroxy-3,5-di-tert.butylbenzoyloxy)-3,4-di-tert.butylbenzamide
16. 1,6-di-[N-acetyl-N-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl)-amino]hexane
17. di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yloxy)-hexane-1,6-dicarbamate
18. 1-acetoxy-4-(N-acetyl-N-n-dodecylamino)-2,2,6,6-tetramethylpiperidine
19. di-(1-propionoxy-2,2,6,6-tetramethylpiperidin-4-yl) succinate
20. di-(4-n-octadecanoyloxy-2,2,6,6-tetramethylpiperazin-1-yl) oxalate
21. 1,4-di-(2-ethylhexanoyloxy)-2,2,6,6-tetramethylpiperidine
22. di-(1-benzoyloxy)-2,2,6,6-tetramethylpiperidin-4-yl) sebacate
23. 1-benzoyloxy-4-(N-n-butyl-N-benzoylamino)-2,2,6,6-tetramethylpiperidine
24. 1-(1-benzoyloxy)-2,2,6,6-tetramethylpiperidin-4-yl)-azepin-2-one
25. [1-benzoyloxy-1'-benzyloxy-di-(2,2,6,6-tetramethylpiperidin-4-yl)]-isophthalate
26. 1,4-di-(4-hydroxy-3,5-di-tert-butylbenzoyloxy)-2,2,6,6-tetramethylpiperidine
27. n-butyl-(4-benzoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) carbonate
28. 1-carbamoyloxy-4-benzoyloxy-2,2,6,6-tetramethylpiperidine
29. di(1-carbamoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) sebacate
30. di-(1-n-butylcarbamoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) 2,2-diethylmalonate
31. di-(4-benzoyloxy-2,2,6,6-tetramethylpiperidin-1-yl)-2,4,4-trimethylhexane-1,6-dicarbamate
32. di-(1-carbamoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) n-butylmalonate
33. di-(1-n-butylcarbamoyloxy-2,2,6,6,-tetramethylpiperidin-4-yl) phthalate
34. di-(1-n-butylcarbamoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) butylmalonate
35. di-(1-phenylcarbamoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) sebacate.

The coating compositions of this invention may be ambient curable or acid catalyzed hot curable systems, depending from the type of binder resins of the composition.

Resins for ambient curable coatings may be, for example, alkyd resins, thermoplastic acrylic resins, acrylic

alkyd resins, polyurethane resins, or polyester resins, said resins may be modified with silicons, isocyan ates, epoxides, isocyanurates, ketimines or oxazolidines. The resins may be esters of cellulose such as nitrocellulose or cellulose acetobutyrate or the resins may be epoxide resins hardenable with polyamines or other hardeners.

Applicable alkyd, acrylic, polyester und epoxide resins are described in S. Paul's "Surface Coatings: Science and Technology" (1985) at pages 70-130. The unmodified and modified alkyd resins which can be stabilized in accordance with the invention, are the conventional resins which are used in trade sales, maintenance and automative refinish coatings. For example, such coatings are based on alkyd resins, alkyd/acrylic resins and alkyd/silicon resins optionally crosslinked by isocyanates or epoxy resins.

In addition various acrylic lacquer coating compositions are disclosed in U.S. 4,168,249. Other acrylic/alkyd resins with polyisocyanate additives are disclosed in U.S. 4,471,083; and acrylic resins containing either pendant amino ester groups or glycidyl groups are described in U.S. 4,525,521.

Resins for acid catalyzed thermosetting coatings may be, for example, hot crosslinkable acrylic, polyester, polyurethane, polyamide or alkyd resins. This implies mixtures of those resins or mixtures with cross-linking agents such as melamine resins.

The acrylic resin lacquers are the conventional acrylic resin stoving lacquers or thermosetting resins including acrylic/melamine systems which are described, for example, in H. Kittel's "Lehrbuch der Lacke und Beschichtungen", Vol. 1, Part 2, on pages 735 and 742 (Berlin 1972), "Lackkunstharze" (1977), by H. Wagner und H.F. Sarx, on pages 229-238, and in S. Paul's "Surface Coatings: Science and Technology" (1985).

The poplyester lacquers are the conventional stoving lacquers described e.g. in H. Wagner and H.F. Sarx, op. cit., on pages 86-99.

The alkyd resin lacquers which can be stabilized in accordance with the invention, are the conventional stoving lacquers which are used in particular for coating automobiles (automobile finishing lacquers), for example lacquers based on alkyd/melamine resins and alkyd/acrylic/melamine resins (see H. Wagner and H.F. Sarx, op. cit., pages 99-123). Other crosslinking agents include glycoluril resins, blocked isocyanates or epoxy resins.

In their industrial uses, enamels with high solids content based on crosslinkable acrylic, polyester, urethane or alkyd resins are cured with an additional acid catalyst. Light stabilizers containing a basic nitrogen group are generally less than satisfactory in this application. Formation of a salt between the acid catalyst and the light stabilizer leads to incompatibility or insolubility and precipitation of the salt and to a reduced level of cure and to reduced light protective action and poor resistance to moisture.

The ambient curable coatings are well as the acid catalyzed hot curable coatings stabilized in accordance with the invention are suitable both for metal finish coatings and solid shade finishes, especially in the case of retouching finishes. The lacquers stabilized in accordance with the invention are preferably applied in the conventional manner by two methods, either by the single-coat method or by the two-coat method. In the latter method, the pigment-containing base coat is applied first and a covering coat of clear lacquer applied over it.

The amount of hindered amine derivative employed is 0.1 to 10 % by weight, based on the solvent-free binder, preferably 0.5 to 5 % by weight. The binders can be dissolved or dispersed in customary organic solvents or in water or can be solvent-free.

When used in two-coat finishes, the hindered amine derivative can be incorporated in the clear coat or both in the clear coat and in the pigmented base coat. In the manufacture of acrylic modified alkyd resins or acrylic resins, polymerizable hindered amine derivatives can be polymerized into the resin. The incorporation into the lacquer binder can also, however, be effected via polycondensation in the manufacture of the alkyd or polyester resins. In these cases, there is the additional advantage that the light stabilizers cannot be removed by extraction or migration so that their action is very prolonged.

To attain maximum light stability, the concurrent use of other conventional light stabilizers can be advantageous. Examples are UV absorbers of the benzophenone, benztriazole, acrylic acid derivative, or oxalanilide type, or aryl-s-triazines or metal-containing light stabilizers, for example organic nickel compounds. In two-coat systems, these additional light stabilizers can be added to the clear coat and/or the pigmented base coat.

If such combinations are employed, the sum of all light stabilizers is 0.2 to 20 % by weight, preferably 0.5 to 5 % by weight, based on the film-forming resin.

Examples of the UV absorbers which may be used in the instant compositions in conjunction with the aforementioned hindered amine compounds are:

(a) 2-(2′-Hydroxyphenyl)-benzotriazoles, for example the 5′-methyl-, 3′,5′-di-tert-butyl-, 5′-tert-butyl-, 5′-(1,1,3,3-tetramethylbutyl)-, 5-chloro-3′,5′-di-tert-butyl-, 5-chloro-3′-tert-butyl-5′-methyl-, 3′-sec-butyl-5′-tert-butyl-, 4′-octoxy-, 3′,5′-di-tert-amyl derivative.

(b) 2-Hydroxy-benzophenones, for example, the 4-hydroxy-, 4-methoxy-, 4-octoxy-, 4-decyloxy-, 4-dodecyloxy-, 4-benzyloxy, 4,2′,4′-trihydroxy-and 2′-hydroxy-4,4′-dimethoxy derivative.

(c) Acrylates, for example, alpha-cyano-β,β-diphenyl-acrylic acid ethyl ester or isoctyl ester, alpha-carbomethoxy-cinnamic acid methyl ester, alpha-cyano-β-methyl-p-methoxy-cinnamic acid methyl ester or butyl ester, alpha-carbomethoxy-p-methoxy-cinnamic acid methyl ester, N-(β-carbomethoxy-β-cyanovinyl)-2-methyl-indoline.

(d) Nickel compounds, for example, nickel complexes of 2,2′-thiobis-[4-(1,1,3,3-tetramethylbutyl)-phe-

nol], such as the 1:1 or 1:2 complex, optionally with additional ligands such as n-butylamine, triethanolamine or N-cyclohexyl-di-ethanolamine, nickel dibutyldithiocarbamate, nickel salts of 4-hydroxy-3,5-di-tert-butylbenzylphosphonic acid monoalkyl esters, such as of the methyl, ethyl or butyl ester, nickel complexes of ketoximes such as of 2-hydroxy-4-methyl-phenyl undecyl ketonoxime, nickel complexes of 1-phenyl-4-lauroyl-5-hydroxy-pyrazol, optionally with additional ligands.

(e) Oxalic acid diamides, for example 4,4′-di-octyl-oxyoxanilide, 2,2′-di-octyloxy-5,5′-di-tert-butyl-oxanilide, 2,2′-di-dodecyloxy-5,5′-di-tert-butyl-oxanilide, 2-ethoxy-2′-ethyl-oxanilide, N-N′-bis-(3-dimethylaminopropyl)-oxalamide, 2-ethoxy-5-tert-butyl-2′-ethyl-oxanilide and its mixture with 2-ethoxy-2′-ethyl-5,4′-di-tert-butyl-oxanilide, and the mixtures of ortho- and paramethoxy- as well as of o-and p-ethoxy-disubstituted oxanilides.

(f) Hydroxyphenyl-5-triazines such as 2,6-bis-(2,4-di-methylphenyl)-4-(2-hydroxy-4-octyloxyphenyl)-s-triazine or the corresponding 4-(2,4-dihydroxyphenyl) derivatives.

Of particular value in the instant compositions are the benzotriazoles of high molecular weight and low volatility such as 2-[2-hydroxy-3,5-di(alpha,alpha-dimethylbenzyl)-phenyl]-benzotriazole, 2-(2-hydroxy-3,5-di-tert-octylphenyl]-benzotriazole, 2-(2-hydroxy-3-alpha,alpha-dimethylbenzyl-5-tert-octylphenyl)-benzotriazole, 2-(2-hydroxy-3-tert-octyl-5,alpha,alpha-dimethylbenzylphenyl)-benzotriazole, 2-(2-hydroxy-3,5-di-tert-amylphenyl)-benzotriazole, 2-[2-hydroxy-3-tert.butyl-5-(2-omega-hydroxy-octa(ethyleneoxy)-carbonyl)-ethylphenyl]-benzotriazole, 2-[2-hydroxy-3-tert.butyl-5-(2-octyloxycarbonyl-ethyl)-phenyl]-benzotriazole, 2-(2-hydroxy-3-sec.dodecyl-5-methyl-phenyl)-benzotriazole, hexamethylene di[β-(3-tert.butyl-4-hydroxy-5-[2-benzotriazolyl]-phenyl)-propionate] and the 5-chloro compounds corresponding to each of the above named benzotriazoles.

Most preferably the benzotriazoles useful in the instant compositions are 2-[2-hydroxy-3,5-di(alpha,alpha-dimethylbenzyl)-phenyl]-benzotriazole and 2-[2-hydroxy-3-tert.butyl-5-(2-omega-hydroxy-octa(ethyleneoxy)-carbonyl)-ethylphenyl]-benzotriazole.

Further ingredients which the enamels or coatings can contain are antioxidants, for example those of the sterically hindered phenol derivative type, phosphorus compounds, such as phosphites, phosphines or phosphonites, conventional hindered amine light stabilizers, plasticizers, levelling assistants, hardening catalysts, thickeners, dispersants or adhesion promoters.

The stabilizers are needed to impart greater retention of durability to the cured enamels (as measured by 20° gloss, distinction of image, cracking or chalking); the enamel should not yellow on curing and further color change on exposure to light should be minimized; the stabilizers should be soluble in the organic solvents normally used in coating applications such as methyl amyl ketone, xylene, n-hexyl acetate, alcohol and the like.

The instant hindered amine light stabilizers substituted on the hindered N-atom by a acyloxy group fulfill each of these requirements and provide individually or in combination with a UV-absorber outstanding light stabilization protection to the cured coatings.

The following examples describe the inventive use of substituted hindered amine derivatives in various ambient curable and acid catalyzed hot curable resin systems. Parts and percentages are by weight.

Example 1 Stabilization of a Tung Oil Phenolic Varnish

Pieces of 1.27 cm x 30.32 cm x 30.48 cm western red cedar panels having a fine radial cut are used to test a commercially available tung oil phenolic varnish (supplied by McCloskey). One half of each panel is coated with two coats of the unstabilized varnish. An equal amount of varnish containing 5 % (by weight based on resins solids) of light stabilizers is applied to the other half of the panel in two coats. After storage for 2 weeks at ambient temperature, the wood panels are exposed outdoors at an angle of 45°S for a period of 8 months. The 60° gloss of each half of the panel is measured at the top, middle, and bottom portion of the panel and averaged (ASTM D 523). Due to the lack of homogeneity of wood substrates, the gloss retention of the same varnish tends to differ slightly from panel to panel. Thus, the application of an unstabilized control varnish to every panel allows for a more meaningful measurement of the improvement in gloss due to the presence of the light stabilizer.

60° Gloss Retention (%)

Compound	Unstabilized	Stabilized	Gloss Improvement
3	24.2	46.7	22.5
A	28.9	39.0	10.1

A = bis(1,2,2,6,6-pentamethyl-4-piperidinyl)sebacate

Example 2 Stabilization of a White Two-Component Polyester Urethane Gloss Enamel

A white polyurethane was formulated as shown below.

Component I	Parts
Desmophen® 670-90 (polyester polyol from Mobay Corp.)	132.4
Titanium Dioxide	198.6
Cellosolve Acetate	98.9
Sand Mill	
Desmophen® 670-90	94.98
Flow Aid	0.28
Tertiary Amine	0.015
Cellosolve Acetate	332.6
Component II	
Desmodur® N-100 (polyisocyanate from Mobay Corp.)	93.9
Cellosolve Acetate	58.7

This material is spray applied at a dry film thickness of 40-50 µm onto Bonderite 40 cold rolled steel panels that have been previously primed with a commercial epoxy polyamide maintenance primer (Sherwin-Williams Tile Clad II). Prior to application, the indicated amounts of hindered amine derivatives (based on resin solids) are added to the paint. After ambient storage for a period of two weeks, three panels of each formulation are exposed outdoors at an angle of 45°S for a period of 9 months. Thereafter, the 20° gloss retention is determined (ASTM D 523-80) at the top, middle and bottom portions of each panel. Thus, the average values for nine gloss retention measurements for each triplicate set of panels are reported below.

Additive	Conc. (% by weight)	20° Gloss Retention (%)
A	1	55
3	1	67
26	1	69

Example 3 Stabilization of a Medium Oil Alkyd Enamel

A medium oil alkyd enamel pigmented with non-leafing aluminum pigment and tinted light blue is stabilized with the indicated amounts of ultraviolet light absorber and hindered amine derivative, and then spray applied onto cold rolled steel panels primed with an epoxy primer. After the coating is allowed to cure at room temperature for 2 weeks, the panels are exposed for accelerated weathering in a Xenon Arc Weatherometer for 840 hours. The 20° gloss values of the panels are determined before and after exposure and indicated below in terms of % gloss retention.

Additive	Conc. (% by weight)	20° Gloss Retention (%)
B/A	3/2	18.9
B/1	3/2	26.4

B = 2-[2-hydroxy-3-tert.butyl-5-(2-(omega-hydroxy-octa-(ethyleneoxy)-carbonyl-ethylphenyl]-benzotriazole

Example 4 Stabilization of an Acrylic Alkyd Crosslinked with an Aliphatic Isocyanate Refinish Enamel

A silver metallic acrylic alkyd enamel hardened with an aliphatic isocyanate is stabilized with the indicated amounts of ultraviolet light absorber and hindered amine derivative (by weight on total resin solids) and then spray applied onto Bonderite 40 panels primed with a black alkyd primer. After the coatings are aged at ambient temperature for 2 weeks, the panels are exposed in a Xenon Arc Weatherometer for 1550 hours. The

20° gloss values of the unexposed and exposed panels are determined and reported below in terms of % gloss retention.

Additive	Conc. (% by weight)	20° Gloss Retention (%)
B/A	3/2	20.7
B/3	3/2	25.7
B/22	3/2	24.3

Example 5

The following alkyd paint formulations are prepared.

	White(W)	Yellow(Y)	Blue(B)
Aroplaz® 1445 M-50 (alkyd resin from NL industries)	45.0	57.73	58.27
TiO_2	45.0	20.82	4.26
Irgalite® GS (yellow pigment from CIBA-GEIGY)	-	7.3	-
Phthalocyanine blue	-	-	1.41
Ketoxime (antiskinning agent)	0.17	0.07	0.014
Ionic antifloat compound	-	-	0.05
24 % Pb as naphthenate	0.94	1.30	1.30
6 % Co as naphthenate	0.36	0.50	0.50
6 % Mn as naphthenate	0.45	0.60	0.60
Xylene	35.0	48.4	45.1
Mineral Spirits	60.0	64.49	64.11

The formulations are stabilized with the indicated materials in the indicated concentrations (by weight on total resin solids) and sprayed onto cold rolled steel panels primed with an electrocoated epoxy primer. The coating is allowed to cure overnight at room temperature and the panels are then exposed in Florida at an angle of 45° South. 60° gloss, distinciton of image (DI) (Hunter Associates Apparatus) and color change based on Yellowness Index values are determined and tabulated below.

Additive	Conc.(% by wt.)	Paint	Florida Exposure (Months)										
			60° Gloss				DI				Color Change		
			0	9	12	15	0	9	12	15	9	12	15
-	-	W	61	30	29	30	63	6	18	25	3.0	2.7	4.0
B/A	1.5/1.5	W	61	28	27	35	55	8	7	19	4.2	4.4	5.4
B/3	1.5/1.5	W	63	43	34	43	80	52	24	38	3.5	3.9	4.8
-	-	Y	81	30	15	14	84	78	12	7	11.0	12.0	13.0
B/A	1.5/1.5	Y	83	41	25	21	78	58	36	12	8.9	11.0	12.0
B/3	1.5/1.5	Y	83	50	31	25	82	73	60	24	8.0	10.0	12.0
-	-	B	86	33	15	11*	84	61	10	0	7.6	11.0	10.0
B/A	1.5/1.5	B	83	63	58	44	80	69	68	59	2.0	2.6	3.3
B/3	1.5/1.5	B	85	63	54	45	86	79	78	64	2.0	4.0	4.3

* = cracking observed

Example 6 Stabilization of a Thermoplastic Acrylic Lacquer

A commercially available light blue metallic thermoplastic acrylic lacquer is stabilized with 2 % each of UV absorber and hindered amine (by weight on total resin solids) and then spray applied onto Bonderite 40 panels primed with an alkyd primer. After storage at ambient temperature for 2 weeks, the panels are exposed in an Xenon Arc Weatherometer for 1250 hours. The 20° gloss retention of the panels are reported below.

Additive	20° Gloss Retention (%)
C/A	11
C/3	29
C/19	35
C/29	40

C = 2-(2-hydroxy-3,5-di(alpha,alpha-dimethyl-benzyl)phenyl)-benzotriazole

Example 7: Stabilization of a High Solids Acid-catalyzed, Thermoset Acrylic Resin Enamel

A high solids (50 % by weight) thermoset acrylic resin enamel, catalyzed by 0.5 % by weight of p-toluenesulfonic acid, based on the film-forming resin, is stabilized by the addition of various derivatives of the instant invention.

The high solids thermoset acrylic resin enamel formulation (Acryloid® AT 400 from Rohm and Haas) is based on hydroxyethyl methacrylate, methyl methacrylate, styrene, butyl acrylate and butyl methacrylate and a melamine curing agent.

Pieces of steel sheeting 4" x 12" (9.16 cm x 30.48 cm), coated with a primer based on polyester/epoxy resin, are then coated with a TiO_2-pigmented base coat on a binder of 70 % of monomers such as hydroxyethyl acrylate, styrene, acrylonitrile, butyl acrylate and acrylic acid with 30 % of a melamine resin and an acid catalyst and finally with the clear finishing enamel. The base coat is sprayed onto the sheet to a thickness of about 0.8 mil (0.02 mm), air dried for 3 minutes and baked at 121° C for 10 minutes. The clear finishing enamel coat is then wire-coated onto the sheet to a thickness of about 0.06 mm. After 15 minutes air-drying, the coated sheets are baked for 30 minutes at 82° C.

The stabilizers under test are added to the thermoset acrylic resin finishing enamel in concentrations of 2 % by weight, before the enamel is coated onto the base coated sheet.

Gardner yellowness index color values are determined on the baked sample and on the sample after having been subjected to an additional 30 minutes baking at 82° C.

In addition, after storage for 3 weeks in an air-conditioned room (23° C/50 % relative humidity), the coated

sheets are subjected to weathering for 150 hours according to test method ASTM G-53/77 in a QUV exposure apparatus. In this apparatus, the samples are subjected to weathering in repreated cycles for 4 hours in a humid atmosphere at 50°C and then for 8 hours under UV light at 70°C. Equivalent samples are also subjected to accelerated weathering for 2625 hours in a Xenon arc (6500 watt) Weatherometer. The samples are exposed to repeated cycles for 102 minutes to the light source, then for 18 minutes to a water spray and light at 60°C. The Gardner color value is also determined for each sample after QUV and Zenon exposure.

Compound	Orig. Color	Color Change		
		Overbake	QUV	Xenon
Unstabilized	1.0	0.8	23.0	24.5
3	1.2	0.8	13.1	3.9
9	1.2	0.6	15.0	4.6
12	1.1	0.3	18.1	4.8
26	3.3	0.8	12.4	1.8
28	1.0	0.3	15.3	3.8

Example 8

The samples of Example 7 are also evaluated on the basis of Knoop Hardness (ASTM D-1474-68) on the baked and overbaked samples; distinction of image (DI), Hunter Associates Apparatus; 20° gloss (ASTM D-523-80); and cracking based on visual observation on a scale of 0-8 with "0" reflecting absence of cracking and "8" relecting excessive cracking and "a", "b", "c" designations for crack depth with "a" relecting shallow cracks and "c" relecting deepest cracks.

Compound	Knoop Hardness		QUV Exposure							
			20° Gloss			DI			Cracks	
	Bake	Overbake	575 hrs.	750	900	575	750	900	750	900
Unstabilized	5.5	8.0	13	11	-	9	4	-	8c	-
3	5.5	8.0	84	70	29	90	85	12	0	3a
9	5.5	8.0	85	56	12	90	62	7	5a	6a
12	4.0	7.0	70	55	10	42	13	8	3a	4a
26	5.5	8.0	85	44	8	92	51	12	7a	7a
28	4.0	6.5	90	66	17	95	77	3	0	7a

These data illustrate a pattern of greater retention of gloss and DI and longer absence of severe cracking after the indicated exposure conditions.

Example 9

The thermoset acrylic enamel of Example 7 is formulated to include 2 % by weight of a benzotriazole UV-absorber and 1.5 %, by weight, of the hindered amine light stabilizer. The enamel is coated over a white base coat or over a silver metallic base coat. Baking is conducted for 30 minutes at 121°C.

The coated panels are exposed in the QUV exposure apparatus and 20° gloss and distinction of image (DI) values are determined.

A) With white base coat

Compound	Hours of QUV Exposure					
	20° Gloss			DI		
	0	1421	2106	0	1421	2106
Unstabilized	93	(655 hrs)*	-	88	(655 hrs)*	-
Compound C	94	9	3*	89	15	3*
C + 3	93	64	16*	89	81	47*
C + 10	93	102	90*	84	82	59*
C + 11	93	80*	-	91	70*	-
C + 12	93	73	27*	92	88	57*
C + 16	93	92	62*	88	81	58*
C + 17	92	67	12*	91	85	45*
C + 18	94	55	7*	89	80	21*
C + 21	92	69*	-	82	57*	-

* = cracking observed

Compound C = 2-[2-hydroxy-3,5-di-(alpha,alpha-dimethylbenzyl)phenyl]-benzotriazole

B) With silver metallic base coat

Compound	Hours of QUV Exposure									
	20° Gloss					DI				
	0	674	916	1118	1377	0	674	916	1118	1377
Unstabilized	92	2*	-	-	-	87	2	-	-	-
Compound C	90	51	5	3	2.2*	84	65	6	5	-2.5
C + 3	95	86	53	18	4.8*	88	79	78	48	28
C + 10	94	85	44	22*	-	88	78	48	13	-
C + 11	92	82	43	27*	-	87	77	36	13	-
C + 12	95	83	59	21	3*	88	87	80	46	-1.8
C + 16	92	93	72	32*	-	90	88	75	28	-
C + 17	93	82	60	33	8*	85	83	80	59	29
C + 18	93	85	40	9	3*	89	86	67	44	20
C + 21	91	89	64	29*	-	71	68	54	28	-

Example 10

Two thermoset acrylic enamels are formulated to include 2 %, by weight, of a benzotriazole UV-absorber and 1 %, by weight of a hindered amine light stabilizer as described in Example 9,

The thermoset acrylic enamels are based on a binder of 70 % of a copolymer from hydroxyethyl acrylate, styrene, acrylonitrile, butyl acrylate and acrylic acid and 30 % of a melamine resin and an acid catalyst (0.5 % of p-toluenesulfonic acid).

Pieces of steel sheeting (9 x 30 cm), coated with a primer based on polyester/epoxy resin, are coated with the white base coat and finally with the clear finishing enamel. The base coat is sprayed onto the sheet to a thickness of about 0.02 mm and air dried for 3 minutes. The clear finishing enamel coat is then sprayed onto the sheet to a thickness of about 0.038 mm. After 15 minutes air-drying, the coated sheets are baked for 30 minutes at 121°C. QUV evaluations are then conducted.

Compound	Test	Hours of QUV Exposure					
		0	1010	1270	1594	1826	2036
Unstabilized	20° Gloss	89	25	23*	-	-	-
	DI	92	5	4	-	-	-
C + 14	20° Gloss	91	91	89	82	48	34*
	DI	98	90	86	87	47	45

Example 11

A white polyester/melamine based oil-free alkyd coil coating is utilized in this example. The fully formulated point is applied over a primed steel sheet using a wire wound rod to give a 0.015-0.020 mm dry film. The panels are baked for 90 seconds at 218°C. The baked panels are removed from the oven and are immediately quenched in water. The coated panels are exposed in a Carbon Arc Weatherometer, a Xenon Arc Weatherometer, and in South Florida at an angle of 45°S to the sun. 20° gloss values are determined.

	Exposure Time	Unstabilized	2 %, by weight Compound
Carbon Arc Weatherometer	0 (Hrs.)	78	81
	112	69	78
	262	15	45
	337	11	37
	447	14	31
Xenon Arc Weatherometer	0 (Hrs.)	78	81
	150	75	83
	378	63	75
	709	11	23
	954	5	12
Florida 45°S Direct Weather	0 (Mos.)	78	81
	6	76	79
	12	39	60

Example 12

The thermoset acrylic enamel of Example 10 including 0.5 % of p-toluenesulfonic acid is formulated to include varying concentrations of a benzotriazole UV-absorber and a hindered amine light stabilizer of the invention. The enamel is coated over a silver metallic base coat pursuant to the procedure in Example 10 and baking is conducted for 30 minutes at 121°C bake temperature.

The coated panels are exposed in the QUV exposure apparatus and the time to 50 % loss of 20° gloss is determined.

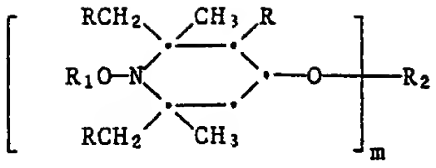
Compound	Conc. (%, by weight)	Time to 50 % loss of 20° Gloss (hours)
unstabilized	-	900
B/1	3.5/1.5	3600
B/2	3.5/1.5	4200
B/23	3.5/1.5	3200
B/26	3.5/1.5	3700
B/32	3.5/1.5	3900
B/33	3.5/1.5	4200
B/35	3.5/1.5	4800
unstabilized	-	1000
B/1	3/1	3800
B/2	3/1	3400
B/4	3/1	3950
B/19	3/1	4000
B/22	3/1	3700
B/25	3/1	3700

Compound
B = 2-[2-hydroxy-3-tert.butyl-5-(2-(omega-hydroxy-octa(ethyleneoxy)carbonyl)-ethylphenyl)]-benzotriazole

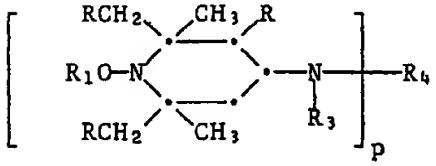
The date in these examples clearly illustrate the beneficial performance characteristics of the compositions of this invention.

Various N-acyloxy hindered amine derivatives have been disclosed. N-Acetoxy derivatives prepared from the N-hydroxy starting material are described in Kurumada et al., J.Polym.Sci., Polym.Chem. Ed. 22, 277-81 (1984). Felder et al., Helv.Chim.Acta. 63, 132 (1980) teach the preparation of an N-phenylacetoxy derivative. The bis(N-acetoxypiperidyl) sebacate has been disclosed in Carlsson et al, Polym.Science Technol. 26, 35-47 (1984). Finally US 4,472,547 discloses various N-piperidyl lactam compounds such as an N-benzoyloxy hindered amine containing a 4-(2-oxo-hexamethyleneimine) substituent as light stabilizers for polyolefins and other organic polymers.

A further object of the invention are the new N-acyloxy hindered amine compounds having one of formulae A' to N'



(A')



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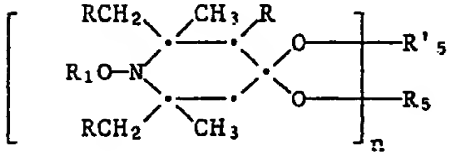

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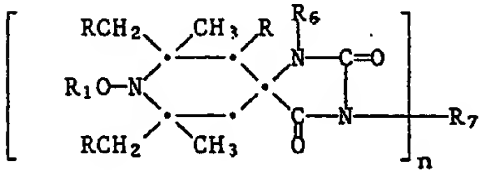

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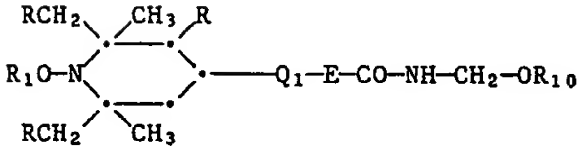

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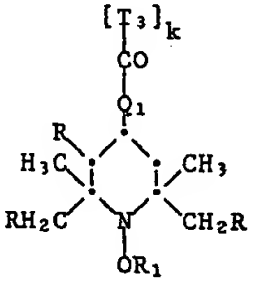

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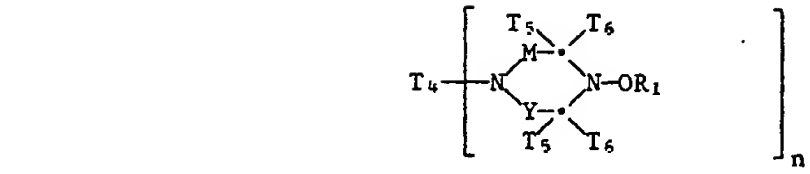

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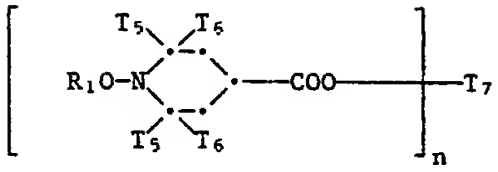
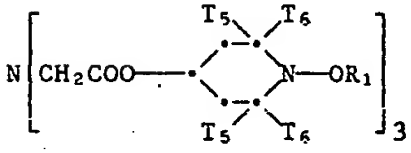
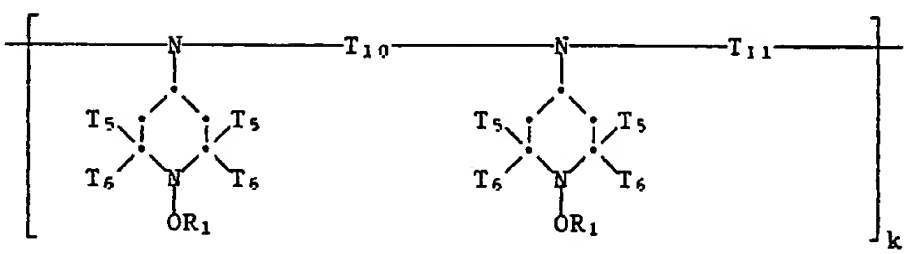


(H')



(I')



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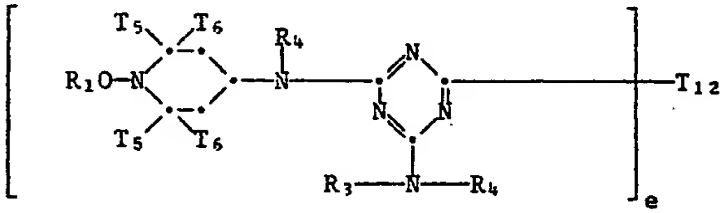

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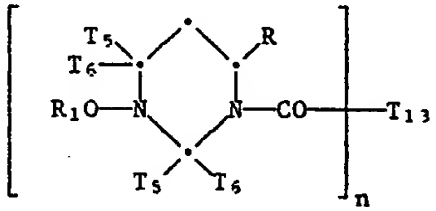
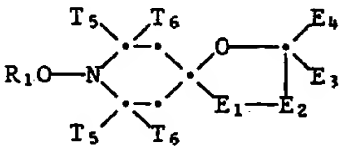
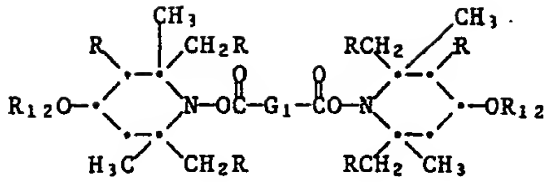


(L')



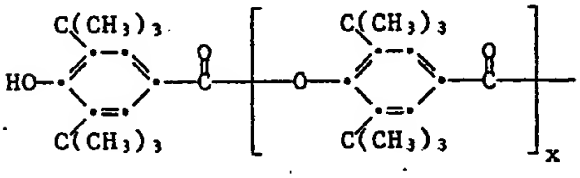
(M')



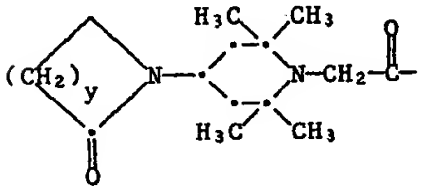
(N')

wherein
R is hydrogen or methyl, preferably hydrogen;
R_1 is a group

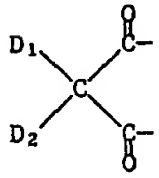
wherein D is C_1-C_{18} alkyl, C_1-C_{18} alkoxy, phenyl, phenyl substituted by hydroxy, C_1-C_4 alkyl or C_1-C_4 alkoxy, or amino or amino mono- or disubstituted by C_1-C_{12} alkyl or phenyl; preferably D is C_1-C_{12} alkyl, C_1-C_4 alkoxy, phenyl, amino, C_1-C_{12} alkylamino or phenylamino;
m is 1-4,
when m is 1,
R_2 is a group



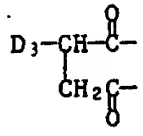
wherein x is 0 or 1, or R_2 is a group



wherein y is 2-4;
when m is 2 and D is alkyl,
R_2 is C_1-C_{12} alkylene, C_4-C_{12} alkenylene, xylylene, a divalent acyl radical of a cycloaliphatic, araliphatic or aromatic dicarboxylic acid having 8-14 C atoms or of an aliphatic, cycloaliphatic or aromatic dicarbamic acid having 8-14 C atoms; or R_2 is a group



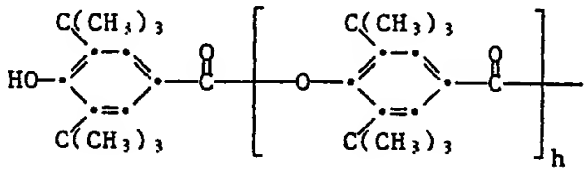
or



wherein D_1 is C_1-C_{18} alkyl, benzyl or 3,5-di-t-butyl-4-hydroxybenzyl, D_2 is D_1 or hydrogen, D_3 is C_1-C_{18} alkyl or C_2-C_{18} alkenyl;
when m is 2 and D is phenyl or substituted phenyl, amino, substituted amino or alkoxy,
R_2 is C_1-C_{12} alkylene, C_4-C_{12} alkenylene, xylylene, a divalent acyl radical of an aliphatic, cycloaliphatic, araliphatic or aromatic dicarboxylic or dicarbamic acid having up to 14 C atoms;

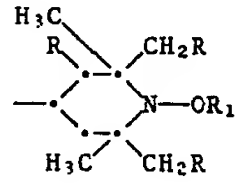
preferably an acyl radical of an aliphatic dicarboxylic acid having 2-18 C atoms, of a cycloaliphatic or aromatic dicarboxylic acid having 8-14 C atoms, or of an aliphatic, cycloaliphatic or aromatic dicarbamic acid having 8-14 C atoms; or R_2 is a group

wherein D_4 and D_5 are independently hydrogen, C_1-C_8 alkyl, benzyl or 3,5-di-t-butyl-4-hydroxybenzyl und D_6 is C_1-C_{18} alkyl or C_2-C_{18} alkenyl;
when m is 3, R_2 is a trivalent acyl radical of an aliphatic, cycloaliphatic, or aromatic tricarboxylic acid having up to 12 C atoms;
when m is 4, R_2 is a tetravalent acyl radical of a saturated or unsaturated aliphatic or aromatic tetracarboxylic acid having up to 18 C atoms including 1,2,3,4-butanetetracarboxylic acid, 1,2,3,4-but-2-enetetracarboxylic acid, and 1,2,3,5-and 1,2,4,5-pentanetetracarboxylic acid;
p is 1, 2 or 3,
R_3 is hydrogen, C_1-C_{12} alkyl, C_5-C_7 cycloalkyl, C_7-C_9 aralkyl, C_2-C_{18} alkanoyl, C_3-C_5 alkenoyl or benzoyl; when p is 1,
R_4 is hydrogen, C_1-C_{18} alkyl, C_5-C_7 cycloalkyl, C_2-C_8 alkenyl unsubstituted or substituted by a cyano, carbonyl or carbamide group, or it is aryl, aralkyl, glycidyl, a group of the formula $-CH_2-CH(OH)-Z$ or $-CONH-Z$ wherein Z is hydrogen, methyl or phenyl;
or R_4 is a group of the formula



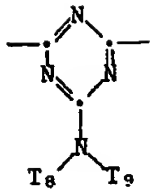
with h as 0 or 1;

or a group of the formula I



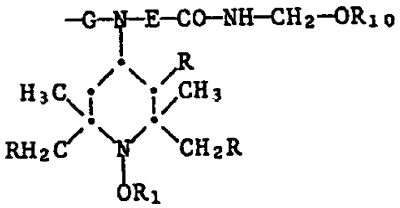
(I);

or R_3 and R_4 together are alkylene of 4 to 6 carbon atoms or 1-oxo alkylene or the divalent acyl radical of an aliphatic or aromatic 1,2- or 1,3-dicarboxylic acid;
when p is 2,
R_4 is C_1-C_{12} alkylene, C_6-C_{12} arylene, xylylene, a $-CH_2CH(OH)-CH_2-$ group, or a group $-CH_2-CH(OH)-CH_2-O-X-O-CH_2-CH(OH)-CH_2-$ wherein X is C_2-C_{10} alkylene, C_6-C_{15} arylene or C_6-C_{12} cycloalkylene; or, provided that R_3 is not alkanoyl, alkenoyl or benzoyl, R_4 can also be a divalent acyl radical of an aliphatic, cycloaliphatic or aromatic dicarboxylic or dicarbamic acid having up to 14 C atoms, or can be the group -CO- or a group of formula II

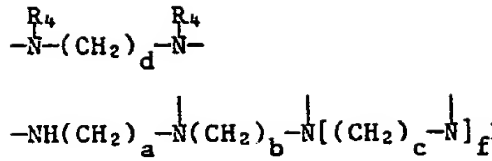


(II)

where T_8 and T_9 are independently hydrogen, alkyl of 1 to 18 carbon atoms, or T_8 and T_9 together are alkylene of 4 to 6 carbon atoms or 3-oxapentamethylene, preferably T_8 and T_9 together are 3-oxapentamethylene;
when p is 3, R_4 is 2,4,6-triazinetriyl;
n is 1 or 2 and when n is 1,
R_5 and R'_5 are independently C_1-C_{12} alkyl, C_2-C_{12} alkenyl, C_7-C_{12} aralkyl, or R_5 is also hydrogen, or R_5 and R'_5 together are C_2-C_8 alkylene or hydroxyalkylene or C_4-C_{22} acyloxyalkylene;
and when n is 2,
R_5 and R'_5 together are $(-CH_2)_2C(CH_2-)_2$;
R_6 is hydrogen, C_1-C_{12} alkyl, allyl, benzyl, glycidyl or C_2-C_6 alkoxyalkyl;
when n is 1,
R_7 is hydrogen, C_1-C_{12} alkyl, C_3-C_5 alkenyl, C_7-C_9 aralkyl, C_5-C_7 cycloalkyl, C_2-C_4 hydroxyalkyl, C_2-C_6 alkoxyalkyl, C_6-C_{10} aryl, glycidyl, a group of the formula $-(CH_2)_t$-COO-Q or of the formula $-(CH_2)_t$-O-CO-Q wherein t is 1 or 2, and Q is C_1-C_4 alkyl or phenyl; and
when n is 2,
R_7 is C_2-C_{12} alkylene, C_6-C_{12} arylene, a group $-CH_2CH(OH)-CH_2-O-X-O-CH_2-CH(OH)-CH_2-$ wherein X is C_2-C_{10} alkylene, C_6-C_{15} arylene or C_6-C_{12} cycloalkylene, or a group $-CH_2CH(OZ')CH_2-(OCH_2-CH(OZ')CH_2)_2-$ wherein Z' is hydrogen, C_1-C_{18} alkyl, allyl benzyl, C_2-C_{12} alkanoyl or benzoyl;
Q_1 is $-N(R_8)-$ or $-O-$;
E is C_1-C_3 alkylene, the group $-CH_2-CH(R_9)-O-$ wherein R_9 is hydrogen, methyl or phenyl, or E is the group $-(CH_2)_3-NH-$ or a direct bond;
R_{10} is hydrogen or C_1-C_{18} alkyl, R_8 is hydrogen, C_1-C_{18} alkyl, C_5-C_7 cycloalkyl, C_7-C_{12} aralkyl, cyanoethyl, C_6-C_{10} aryl, the group $-CH_2-CH(R_9)-OH$ wherein R_9 has the meaning defined above; a group of the formula I or a group of the formula



wherein G can be C_2-C_8 alkylene or C_6-C_{12} arylene; or R_8 is a group $-E-CO-NH-CH_2-OR_{10}$;
Formula F denotes a recurring structural unit of a polymer where T_3 is ethylene or 1,2-propylene, it is the repeating structural unit derived from an alpha-olefin copolymer with an alkyl acrylate or methacrylate; preferably a copolymer of ethylene and ethyl acrylate, and where k is 2 to 100;
T_4 has the same meaning as R_4 when p is 1 or 2,
T_5 is methyl,
T_6 is methyl or ethyl, or T_5 and T_6 together are tetramethylene or pentamethylene, preferably T_5 and T_6 are each methyl,
M and Y are independently methylene or carbonyl, preferably M is methylene, Y is carbonyl and T_4 is ethylene where n is 2;
T_7 is the same as R_7, and T_7 is preferably octamethylene when n is 2;
T_{10} and T_{11} are independently alkylene of 2 to 12 carbon atoms, or T_{11} is a group of formula II;
e is 2, 3 or 4,
T_{12} is a group



where a, b and c are independently 2 or 3, d is 2-10, and f is 0 or 1, preferably a and c are each 3, b is 2 and f is 1;
T_{13} is the same as R_4 with the proviso that T_{13} cannot be hydrogen when n is 1;
E_1 and E_2, being different, each are -CO- or -N(E_5)-, where E_5 is hydrogen, C_1-C_{12} alkyl or alkoxycarbonylalkyl of 4 to 22 carbon atoms, preferably E_1 is -CO- and E_2 is -N(E_5);
E_3 is hydrogen, alkyl of 1 to 30 carbon atoms, phenyl, naphthyl, said phenyl or said naphthyl substituted by chlorine or by alkyl of 1 to 4 carbon atoms, or phenylalkyl of 7 to 12 carbon atoms, or said phenylalkyl substituted by alkyl of 1 to 4 carbon atoms,
E_4 is hydrogen, alkyl of 1 to 30 carbon atoms, phenyl, naphthyl or phenylalkyl of 7 to 12 carbon atoms, or E_3 and E_4 together are polymethylene of 4 to 17 carbon atoms, or said polymethylene substituted by up to four alkyl groups of 1 to 4 carbon atoms, preferably methyl, and
R_{12} is C_2-C_{18} alkanoyl, C_3-C_6 alkenoyl, benzoyl or benzoyl substituted by C_1-C_4 alkyl, halogen or hydroxyl, and G_1 is a direct bond, C_1-C_{12} alkylene, phenylene or -NH-G'-NH wherein G' is C_1-C_{12} alkylene.

In the structures A' to N', if any substituents are C_1-C_{18} alkyl, they are for example methyl, ethyl, n-propyl, n-butyl, sec-butyl, tert-butyl, n-hexyl, n-octyl, 2-ethylhexyl, n-nonyl, n-decyl, n-undecyl, n-dodecyl, n-tridecyl, n-tetradecyl, n-hexadecyl or n-octadecyl. Typical cycloalkyl groups include cyclopentyl and cyclohexyl, while typical aralkyl groups include benzyl, alpha-methyl-benzyl, alpha,alpha-dimethylbenzyl or phenethyl.

If R_2 is a divalent acyl radical of a dicarboxylic acid, it is for example an acyl radical of adipic acid, succinic acid, suberic acid, sebacic acid, phthalic acid, dibutylmalonic acid, dibenzylmalonic acid or (3,5-di-tert-butyl-4-hydroxybenzyl)-malonic acid, or bicycloheptenedicarboxylic acid.

If R_2 is a divalent acyl radical of a dicarbamic acid, it is for example an acyl radical of hexamethylenedicarbamic acid or of 2,4-toluylenedicarbamic acid.

The following compounds are examples of polyalkylpiperidine starting materials useful in making hindered amine derivatives of formula A'. (Depending on the selected preparative procedure).

di-(2,2,6,6-tetramethylpiperidin-4-yl) phthalate
alpha,alpha'-(di-2,2,6,6-tetramethylpiperidyl-4-oxy)-p-xylene
1,4-dihydroxy-2,2,6,6-tetramethylpiperidine
1-acetoxy-4-hydroxy-2,2,6,6-tetramethylpiperidine
di-(1-oxyl-2,2,6,6-tetramethylpiperidin-4-yl) phthalate
1-oxyl-2,2,6,6-tetramethylpiperidin-4-one
(1-hydroxy-2,2,6,6-tetramethylpiperidin-4-yl) stearate
(2,2,6,6-tetramethylpiperidin-4-yl)-[4-(2-oxoazepin-1-yl)-2,2,6,6-tetramethylpiperidin-4-yl] acetate

As C_2-C_{18} alkanoyl, R_3 is for example propionyl, butyryl, octanoyl, dodecanoyl, hexadecanoyl, octadecanoyl, but preferably acetyl; and as C_3-C_5 alkenoyl, R_3 is in particular acryloyl.

If R_4 is C_2-C_8 alkenyl unsubstituted or substituted by a cyano, carbonyl or carbamide group, it is for example 1-propenyl, allyl, methallyl, 2-butenyl, 2-pentenyl, 2-hexenyl, 2-octenyl, 2,2-dicyanovinyl, 1-methyl-2-cyano-2-methoxycarbonyl-vinyl or 2,2-diacetylaminovinyl.

If any substituents are C_2-C_{12} alkylene, they are for example ethylene, propylene, 2,2-dimethylpropylene, tetramethylene, hexamethylene, octamethylene, decamethylene or dodecamethylene.

If any substituents are C_6-C_{12} arylene, they are for example o-, m- or p-phenylene, 1,4-naphthylene or 4,4'-di-phenylene.

As C_6-C_{12} cycloalkylene, X is especially cyclohexylene.

The following compounds are examples of polyalkylpiperidine starting materials useful in making compounds of formula B'.

N,N'-bis-(2,2,6,6-tetramethylpiperidin-4-yl)-hexamethylene-1,6-diamine,
N,N'-bis-(2,2,6,6-tetramethylpiperidin-4-yl)-hexamethylene-1,6-diacetamide,
4-benzylamino-2,2,6,6-tetramethylpiperidine,
N-n-butyl-N-(2,2,6,6-tetramethylpiperidine-4-yl)-4-hydroxy-3,5-di-tert.butylbenzamide,
N,N'-bis-(2,2,6,6-tetramethylpiperidin-4-yl)-N,N'-di-butyl-adipamide,
N,N'-bis-(2,2,6,6-tetramethylpiperidin-4-yl)-N,N'-dicyclohexyl-(2-hydroxypropylene),
N,N'-bis-(2,2,6,6-tetramethylpiperidin-4-yl)-p-xylylenediamine,
4-(3-methyl-4-hydroxy-5-tert-butyl-benzoyl acetamido)-2,2,6,6-tetramethylpiperidine,
alpha-cyano-β-methyl-β-[N-(2,2,6,6-tetramethylpiperidin-4-yl-amino]-acrylic acid methyl ester,
1-acetoxy-4-butylamino-2,2,6,6-tetramethylpiperidine,
1-oxyl-2,2,6,6-tetramethylpiperidin-4-one.

If R_5 and R'_5 together are C_2-C_8 alkylene or hydroxyalkylene or C_4-C_{22} acyloxyalkylene, it is for example ethylene, 1-methyl-ethylene, propylene, 2-ethylpropylene, 2-ethyl-2-hydroxymethylpropylene or 2-ethyl-2-acetoxymethyl-propylene.

The following compounds are examples of polyalkylpiperidine starting materials useful in making the compounds of formula C'.

9-aza-8,8,10,10-tetramethyl-1,5-dioxaspiro[5.5]undecane,
9-aza-8,8,10,10-tetramethyl-3-ethyl-1,5-dioxaspiro[5.5]undecane,
2,2,6,6-tetramethylpiperidine-4-spiro-2'-(1',3'-dioxane)-5'-spiro-5''-(1'',3''-dioxane)-2''-spiro-4'''-(2''',2''',6''',6'''-tetramethylpiperidine).

If any substituents are C_2-C_8 alkoxyalkyl, they are for example methoxymethyl, ethoxymethyl, propoxymethyl, tert-butoxyethyl, ethoxyethyl, ethoxypropyl, n-butoxyethyl, tert-butoxyethyl, isopropoxyethyl or propoxypropyl.

If R_7 is C_3-C_5 alkenyl, it is for example 1-propenyl, allyl, methallyl, 2-butenyl or 2-pentenyl.

As C_7-C_9 aralkyl, R_7 is in particular phenethyl or above all benzyl; and as C_5-C_7 cycloalkyl, R_7 is especially cyclohexyl.

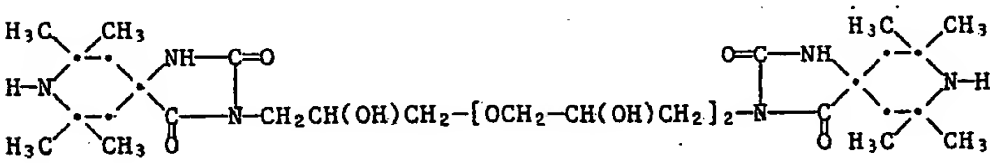
If R_7 is C_2-C_4 hydroxyalkyl, it is for example 2-hydroxyethyl, 2-hydroxypropyl, 3-hydroxypropyl, 2-hydroxybutyl or 4-hydroxybutyl.

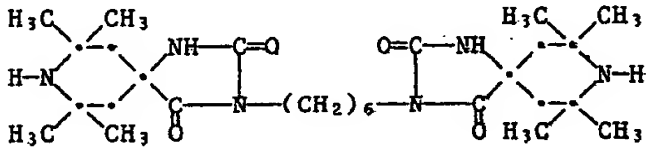
As C_6-C_{10} aryl, the substituents are in particular phenyl, or alpha- or β-naphthyl which is unsubstituted or substituted by halogen or C_1-C_4 alkyl.

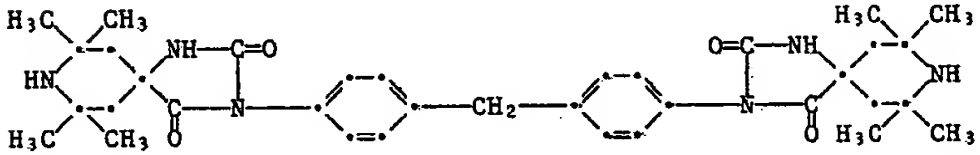
If Z' is C_2-C_{12} alkanoyl, it is for example propionyl, butyryl, octanoyl, dodecanoyl or preferably acetyl.

The following compounds are examples of polyalkylpiperidine starting materials useful in making compounds of formula D'.

3-benzyl-1,3,8-triaza-7,7,9,9-tetramethylspiro[4.5]-decane-2,4-dione,
3-n-octyl-1,3,8-triaza-7,7,9,9-tetramethylspiro[4.5]-decane-2,4-dione,
3-allyl-1,3,8-triaza-1,7,7,9,9-pentamethylspiro[4.5]-decane-2,4-dione,
or the compounds of the following formulae:







As C_5-C_7 cycloalkyl, R_8 is in particular cyclohexyl.

As C_6-C_{10} aryl, R_8 is particularly phenyl, or alpha- or β-naphthyl which is unsubstituted or substituted with halogen or C_1-C_4 alkyl.

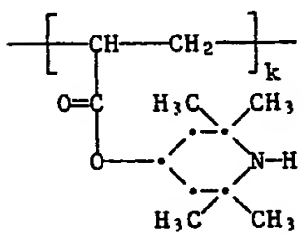
As C_1-C_3 alkylene, E is for example methylene, ethylene or propylene.

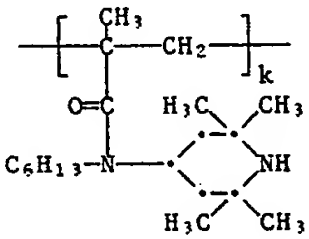
As C_2-C_8 alkylene, G is for example ethylene, propylene, 2,2-dimethylpropylene, tetramethylene or hexamethylene; and as C_6-C_{12} arylene, G is o-, m- or p-phenylene, 1,4-naphthylene or 4,4'-diphenylene.

The following compounds are examples of polyalkylpiperidine starting materials useful in making the compounds of formula E'.

N-hydroxymethyl-N'-2,2,6,6-tetramethylpiperidin-4-yl-urea,
N-methoxymethyl-N'-2,2,6,6-tetramethylpiperidin-4-yl-urea,
N-methoxymethyl-N'-n-dodecyl-N'-2,2,6,6-tetramethylpiperidin-4-yl-urea, and
O-(2,2,6,6-tetramethylpiperidin-4-yl)-N-methoxymethyl-urethane.

When the instant hindered amine derivative is of formula F', the following polymeric compounds are examples of starting materials useful in preparing said derivatives.

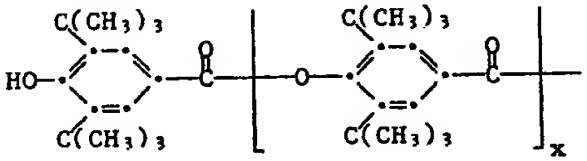




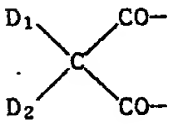
Additional starting hindered amine derivatives include for formula J':

poly-{[6-[(1,1,3,3-tetramethylbutyl)-imino]-1,3,5-triazine-2,4-diyl][2-(1-oxyl-2,2,6,6-tetramethylpiperidyl)-imino]-hexamethylene-4[4-(1-oxyl-2,2,6,6-tetramethylpiperidyl]-imino]}.

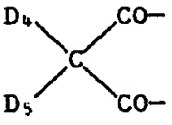
Preferred compounds are the compounds of formulae A', B', D', F', J', K', M' or N', wherein R is hydrogen, R_1 is -CO-D and D is C_1-C_{12} alkyl, C_1-C_4 alkoxy, phenyl, amino, C_1-C_{12} alkylamino or phenylamino, m is 1, 2 or 4, and
when m is 1, R_2 is



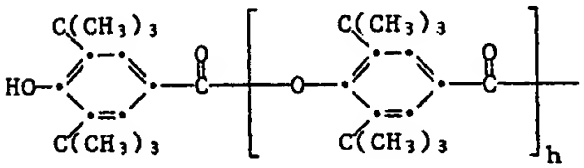
wherein x is 0 or 1,
when m is 2 and D is alkyl,
R_2 is a divalent acyl radical of a cycloaliphatic or aromatic dicarboxylic acid having 8-14 C atoms, or of an aliphatic, cycloaliphatic or aromatic dicarbamic acid having 8-14 C atoms; or R_2 is a group



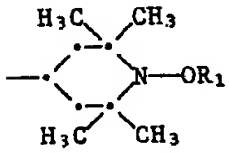
wherein D_1 is C_1-C_4 alkyl or 3,5-di-t-butyl-4-hydroxybenzyl and D_2 is D_1 or hydrogen, and when m is 2 and D is phenyl, amino, alkylamino, phenylamino or alkoxy, R_2 is a divalent acyl radical of an aliphatic, dicarboxylic acid having 2-8 C atoms or of a cycloaliphatic or aromatic dicarboxylic acid having 8-14 C atoms or of an aliphatic, cycloaliphatic or aromatic dicarbamic acid having 8-14 C atoms, or R_2 is a group



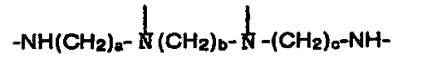
wherein D_4 and D_5 are independently hydrogen, C_1-C_8 alkyl, benzyl or 3,5-di-t-butyl-4-hydroxybenzyl, and when m is 4, R_2 is a tetravalent acyl radical of 1,2,3,4-butanetetracarboxylic acid, 1,2,3,4-but-2-enetetracarboxylic acid, 1,2,3,5- or 1,2,4,5-pentanetetracarboxylic acid;
p is 1, 2 or 3,
R_3 is hydrogen, C_1-C_{12} alkyl, cyclohexyl, C_7-C_9 aralkyl, C_2-C_{18} alkanoyl or benzoyl;
when p is 1,
R_4 is C_1-C_{18} alkyl, cyclohexyl, allyl, benzyl or a group



with h as 0 or 1, or is a group

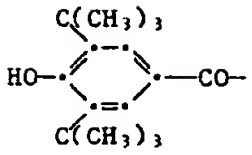


when p is 2,
R_4 is C_1-C_{12} alkylene, C_6-C_{12} arylene, xylylene, a group $-CH_2CH(OH)-CH_2-$, or provided that R_3 is not alkanoyl or benzoyl, R_4 can also be a divalent acyl radical of an aliphatic, cycloaliphatic or aromatic dicarboxylic acid or dicarbamic acid having 6-12 C atoms,
or R_4 is a group of formula II,
where T_8 and T_9 are independently hydrogen or C_1-C_{12} alkyl or T_8 and T_9 together are C_4-C_6 alkylene or 3-oxapentamethylene,
when p is 3, R_4 is 2,4,6-triazinetriyl;
n is 1 or 2 and
R_6 is hydrogen, C_1-C_{12} alkyl, allyl or benzyl,
when n is 1, R_7 is hydrogen, C_1-C_{12} alkyl, allyl, benzyl or cyclohexyl, and when n is 2, R_7 is C_2-C_{12} alkylene;
k is 5 to 50, T_3 is ethylene or propylene and Q_1 i NH or o;
T_5 and T_6 each are methyl,
T_{10} and T_{11} are independently C_2-C_{12} alkylene or T_{11} is a group of formula II,
e is 4 and
T_{12} is

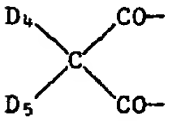


where a, b and c are independently 2 or 3;
E_1 is -CO- and E_2 is $-N(E_5)-$, E_3 and E_4 independently are C_1-C_{12} alkyl or E_3 and E_4 together are C_5-C_{12} polymethylene, and E_5 is hydrogen or C_1-C_{12} alkyl;
G_1 is a direct bond, C_2-C_8 alkylene or -NH-G'-NH- wherein G' is C_2-C_8 alkylene, and R_{12} is C_2-C_{18} alkanoyl or benzoyl.

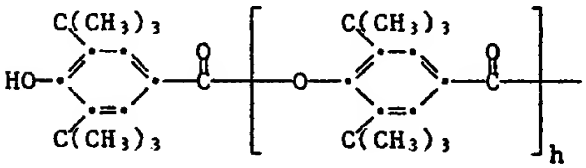
Especially preferred are the compounds of formulae A', B', F', J' or N', wherein R is hydrogen,
R_1 is -CO-D and D is C_1-C_4 alkyl, C_1-C_4 alkoxy, phenyl, amino, C_1-C_4 alkylamino or phenylamino;
m is 1 or 2 and
when m is 1, R_2 is a group



when m is 2 and D is alkyl, R_2 is a divalent acyl radical of a phenylenedicarboxylic acid or is a group -CO-C(D_1)(D_2)-CO- wherein D_1 is C_1-C_4 alkyl or 3,5-di-t-butyl-4-hydroxybenzyl and D_2 is D_1 or hydrogen and when m is 2 and D is alkoxy, phenyl, amino or alkylamino,
R_2 is a divalent acyl radical of an aliphatic or aromatic dicarboxylic acid having 8-10 C atoms or of an aliphatic dicarbamic acid having 8-10 C atoms; or R_2 is a group



wherein D_4 is alkyl or 3,5-di-t-butyl-4-hydroxybenzyl and D_5 is D_4 or hydrogen;
p is 1 or 2, R_3 is C_1-C_4 alkyl, acetyl or benzoyl,
when p is 1, R_4 is C_1-C_{12} alkyl or a group



with h as 0 or 1,
and when p is 2, R_4 is C_4-C_8 alkylene;
k is 5 to 50, T_3 is ethylene and Q_1 is 0;
T_5 and T_6 are methyl,
T_{10} is hexamethylene and T_{11} is a group of formula II
wherein T_8 and T_9 are independently hydrogen or C_1-C_{12} alkyl or T_8 and T_9 together are pentamethylene or 3-oxapentamethylene;
R_{12} is acetyl or benzoyl and G_1 is a direct bond or -NH-(CH_2)$_6$-NH-.

The compounds of formula A' are generally prepared by oxidizing the corresponding hindered amine with an appropriate peroxy compound such as hydrogen peroxide or tert-butyl hydroperoxide in the presence of a metal carbonyl or metal oxide catalyst followed by reduction of the oxyl intermediate formed to the desired N-hydroxy derivative, preferably by catalytic hydrogenation.

Thereafter, the N-acyloxy, N-carbamoyloxy and N-(alkoxyacyl)oxy derivatives are prepared by reacting the N-hydroxy hindered amine with the appropriate acid chloride, anhydride, cyanate, isocyanate or substituted chloroformate (for carbonates). The catalytic hydrogenation can also be conducted in acetic anhydride to prepare the N-acetoxy derivative.

The compounds of formula N' can be prepared analogously by reacting the 1-hydroxy intermediates with an dicarboylic acid derivative or with a diisocyanate.

These reactions are generally conducted at temperatures ranging from 0 to 60° C and in a variety of solvents including toluene and dichloromethane. The acylations with carboxylic acid chlorides are preferably conducted in the presence of an acid acceptor such as triethylamine. The various hindered amine precursors are largely commercially available or can be prepared by methods known in the art.

The derivatives are particularly effective in stabilizing organic materials against the degradative effects of actinic stimuli. Such organic materials include polymeric materials such as the following polymers:

1. Polymers of monoolefins and diolefins, for example polypropylene, polyisobutylene, polybutene-1, polymethylpentene-1, polyisoprene or polybutadiene, as well as polymers of cycloolefins, for instance of cyclopentene or norbornene, polyethylene (which optioanlly can be crosslinked), for example high density polyethylene (HDPE), low density polyethylene (LDPE) and linear low density polyethylene (LLDPE).

2. Mixtures of the polymers mentioned under 1), for example mixtures of polypropylene with polyisobutylene, polypropylene with polyethylene (for example PP/HDPE, PP/LDPE) and mixtures of different types of polyethylene (for example LDPE/HDPE).

3. Copolymers of monoolefines and diolefines with each other or with other vinyl monomers, such as, for example, ethylene/propylene, linear low density polyethylene (LLDPE) and its mixtures with low density polyethylene (LDPE), propylene/butene-1, ethylene/hexene, ethylene/ethylpentene, ethylene/heptene, ethylene/octene, propylene/isobutylene, ethylene/butene-1, propylene/butadiene, isobutylene/isoprene, ethylene/alkyl acrylates, ethylene/alkyl methacrylates, ethylene/vinyl acetate or ethylene/acrylic acid copolymers and their salts (ionomers) and terpolymers of ethylene with propylene and a diene, such as hexadiene, dicyclopentadiene or ethylidene-norbornene; as well as mixtures of such copolymers and their mixtures with polymers mentioned in 1) above, for example polypropylene/ethylene-propylene-copolymers, LDPE/EVA, LDPE/EAA, LLDPE/EVA and LLDPE/EAA.

3a. Hydrocarbon resins (for example C_5-C_9) and hydrogenated modifications thereof (for example tackyfiers).

4. Polystyrene, poly-(p-methylstyrene), poly-(α-methylstyrene).

5. Copolymers of styrene or α-methylstyrene with dienes or acrylic derivatives, such as, for example, styrene/butadiene, styrene/ acrylonitrile, styrene/alkyl methacrylate, styrene/maleic anhydride, styrene/butadiene/ethyl acrylate, styrene/acrylonitrile/methyl acrylate; mixtures of high impact strength from styrene copolymers and another polymer, such as, for example, from a polyacrylate, a diene polymer or an ethylene/propylene/diene terpolymer; and block copolymers of styrene, such as, for example, styrene/butadiene/ styrene, styrene/ isoprene/styrene, styrene/ethylene/butylene/ styrene or styrene/ethylene/propylene/styrene.

6. Graft copolymers of styrene or α-methylstyrene such as, for example, styrene on polybutadiene, styrene on polybutadiene-styrene or polybutadiene-acrylonitrile; styrene and acrylonitrile (or methacrylonitrile) on polybutadiene; styrene and maleic anhydride or maleimide on polybutadiene; styrene, acrylonitrile and maleic anhydride or maleimide on polybutadiene; styrene, acrylonitrile and methyl methacrylate on polybutadiene, styrene and alkyl acrylates or methacrylates on polybutadiene, styrene and acrylonitrile on ethylene/propylene/diene terpolymers, styrene and acrylonitrile on polyacrylates or polymethacrylates, styrene and acrylonitrile on acrylate/butadiene copolymers, as well as mixtures thereof with the copolymers listed under 5), for instance the copolymer mixtures known as ABS-, MBS-, ASA- or AES-polymers.

7. Halogen-containing polymers, such as polychloroprene, chlorinated rubbers, chlorinated or sulfochlorinated polyethylene, epichlorohydrin homo- and copolymers, polymers from halogen-containing vinyl compounds,as for example, polyvinylchloride, polyvinylidene chloride, polyvinyl fluoride, polyvinylidene fluoride, as well as copolymers thereof, as for example, vinyl chloride/vinylidene chloride, vinyl chloride/vinyl acetate or vinylidene chloride/vinyl acetate copolymers.

8. Polymers which are derived from α,β-unsaturated acids and derivatives thereof, such as polyacrylates and polymethacrylates, polyacrylamide and polyacrylonitrile.

9. Copolymers from the monomers mentioned under 8) with each other or with other unsaturated monomers, such as, for instance, acrylonitrile/butadiene, acrylonitrile/alkyl acrylate, acrylonitrile/alkoxyalkyl acrylate or acrylonitrile/vinyl halogenide copolymers or acrylonitrile/alkyl methacrylate/butadiene terpolymers.

10. Polymers which are derived from unsaturated alcohols and amines, or acyl derivatives thereof or acetals thereof, such as polyvinyl alcohol, polyvinyl acetate, polyvinyl stearate, polyvinyl benzoate, polyvinyl maleate, polyvinyl butyral, polyallyl phthalate or polyallylmelamine; as well as their copolymers with olefins mentioned in 1) above.

11. Homopolymers and copolymers of cyclic ethers, such as polyalkylene glycols, polyethylene oxide, polypropylene oxide or copolymers thereof with bis-glycidyl ethers.

12. Polyacetals, such as polyoxymethylene and those polyoxymethylenes which contain ethylene oxide as a comonomer; polyacetals modified with thermoplastic polyurethanes, acrylates or MBS.

13. Polyphenylene oxides and sulfides, and mixtures of polyphenylene oxides with polystyrene or polyamides.

14. Polyurethanes which are derived from polyethers, polyesters or polybutadienes with terminal hydroxyl groups on the one side and aliphatic or aromatic polyisocyanates on the other side, as well as precursors thereof (polyisocyanates, polyols or prepolymers).

15. Polyamides and copolyamides which are derived from diamines and dicarboxylic acids and/or from aminocarboxylic acids or the corresponding lactams, such as polyamide 4, polyamide 6, polyamide 6/6, 6/10, 6/9, 6/12 and 4/6, polyamide 11, polyamide 12, aromatic polyamides obtained by condensation of m-xylene diamine and adipic acid; polyamides prepared from hexamethylenediamine and isophthalic or/and terephthalic acid and optionally an elastomer as modifier, for example poly-2,4,4,-trimethylhexamethylene terephthalamide or poly-m-phenylene isophthalamide. Further copolymers of the aforementioned polyamides with polyolefins, olefin copolymers, ionomers or chemically bonded or grafted elastomers; or with polyethers, such as for instance, with polyethylene glycols, polypropylene glycols or polytetramethylene glycols. Polyamides or copolyamides modified with EPDM or ABS. Polyamides condensed during processing (RIM-polyamide systems).

16. Polyureas, polyimides and polyamide-imides.

17. Polyesters which are derived from dicarboxylic acids and diols and/or from hydroxycarboxylic acids or the corresponding lactones, such as polyethylene terephthalate, polybutylene terephthalate, poly-1,4-dimethylolcyclohexane terephthalate, poly-[2,2,-(4-hydroxyphenyl)-propane] terephthalate and polyhydroxybenzoates as well as block-copolyether-esters derived from polyethers having hydroxyl end groups.

18. Polycarbonates and polyester-carbonates.

19. Polysulfones, polyether-sulfones and polyether-ketones.

20. Crosslinked polymers which are derived from aldehydes on the one hand and phenols, ureas and melamines on the other hand, such as phenol/formaldehyde resins, urea/formaldehyde resins and melamine/formaldehyde resins.

21. Drying and non-drying alkyd resins.

22. Unsaturated polyester resins which are derived from copolyesters of saturated and unsaturated dicarboxylic acids with polyhydric alcohols and vinyl compounds as crosslinking agents, and also halogen-containing modifications thereof of low inflammability.

23. Thermosetting acrylic resins, derived from substituted acrylic esters, such as epoxy-acrylates, urethane-acrylates or polyester-acrylates.

24. Alkyd resins, polyester resins or acrylate resins in admixture with melamine resins, urea resins, polyisocyanates or epoxide resins as crosslinking agents.

25. Crosslinked epoxide resins which are derived from polyepoxides, for example from bis-glycidyl ethers or from cycloaliphatic diepoxides.

26. Natural polymers, such as cellulose, rubber, gelatine and derivatives thereof which are chemically modified in a polymer-homologous manner, such as cellulose acetates, cellulose propionates and cellulose butyrates, or the cellulose ethers, such as methylcellulose; rosins and their derivatives.

27. Mixtures of polymers as mentioned above, for example PP/EPDM, Polyamide 6/EPDM or ABS, PVC/EVA, PVC/ABS, PVC/MBS, PC/ABS, PBTP/ABS, PC/ASA, PC/PBT, PVC/CPE, PVC/acrylates, POM/thermoplastic PUR, PC/thermoplastic PUR, POM/acrylate, POM/MBS, PPE/HIPS, PPE/PA 6.6 and copolymers, PA/HDPE, PA/PP, PA/PPE.

The instant compounds are added to the polymers in a concentration of 0.05 to 5 % by weight, calculated relative to the material to be stabilized. Preferably, 0.1 to 2.5 % by weight of the stabilizer calculated relative to the material to be stabilized, is incorporated into the latter.

Incorporation can be effected during the polymerization or after polymerization, for example by mixing the compounds and, if desired, further additives into the melt by the methods customary in the art, before or during shaping, or by applying the dissolved or dispersed compounds to the polymer.

Further additives used in combination with the instant compounds may be other stabilizers such as phenolic antioxidants, metal desactivators, phosphites, thiodipropionic diesters, fatty acid salts, UV-absorbers or nickel complex salts. Further additives may be pigments, fillers, plasticizers, flame retardants or antistatica.

In general, the stabilizers of this invention are employed from about 0.05 to about 5 % by weight of the stabilized composition, although this will vary with the particular substrate and application. An advantageous range is from about 0.1 to about 2.5 %.

The compounds are particularly active as light stabilizers in ambient cured and acid catalyzed thermoset coatings or enamels. Since these materials are considerably less basic than conventional hindered amines, they do not inhibit or interfere with cure as is encountered with the conventional hindered amines nor are they interacting. They likewise do not exhibit the color problems encountered with nitroxyl radicals and, unlike N-hydroxy hindered amines, tend to resist air oxidation during handling. Finally, the N-acyloxy hindered amines exhibit greater solubility in the solvents typically utilized in coatings. These areas are further described in a copending application.

The compounds alone or in combination with phenols can further be used in photographic layers as yellow dye light stabilizers, as cyan dye dark stabilizers, as antistain agents in magenta layers (especially for two-equivalent magenta couplers) and as thermal stabilizers for magenta couplers.

The following examples further illustrate the preparation of these compounds.

Example 13 Di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) Phthalate

A mixture of 20.0 g (42 mmol) of di-(1-oxyl-2,2,6,6-tetramethyl-piperidin-4-yl) phthalate, 100 ml of acetic anhydride, and 500 mg of 5 % Pd on C is hydrogenated in a Parr apparatus (50 psi, ambient temperature, 1 hour). Catalyst is filtered and solvent is evaporated. The residue is dissolved in 150 ml of ethyl acetate. The ethyl acetate solution is washed with 5 % aqueous ammonia (2 x 100 ml), water (100 ml), and saturated sodium chloride (100 ml), then dried over magnesium sulfate and concentrated to obtain a crude solid. The crude product is recrystallized from methanol to obtain 14.9 g (64 % yield) of a white crystalline solid, m.p. 172-175°C.

Anal. Calcd. for $C_{30}H_{44}N_2O_8$:
C, 64.3; H, 7.9; N, 5.0.
Found:
C, 64.5; H, 8.2; N, 5.1.

Example 14 Di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) isophthalate

The compound is prepared according to the procedure given for Example 13 utilizing the isophthalate in place of the phthalate, m.p. 98-101°C.

Anal. Calcd. for $C_{30}H_{44}N_2O_8$:
C, 64.3; H, 7.9; N, 5.0.
Found:
C, 62.4; H, 8.0; N, 5.0.

Example 15 Poly-{[6-[(1,1,3,3-tetramethylbutyl)-imino]-1,3,5-triazine-2,4-diyl] [2-(1-acetoxy-2,2,6,6-tetramethylpiperidyl)-imino]-hexamethylene-[4-(1-acetoxy-2,2,6,6-tetramethylpiperidyl]-imino]}

The compound is prepared according to the procedure given for Example 13, utilizing the corresponding 1-oxyl compound
m.p. 110-120°C (glass transition).
Anal. Calcd. for $(C_{39}H_{70}N_8O_4)_n$:
C, 65.5; H, 9.9; N, 15.7.
Found:
C, 65.6; H, 9.4; N, 13.0.

Example 16 Polymer of 1-acetoxy-4-acryloxy-2,2,6,6-tetramethylpiperidine

A solution of 20.0 g (74.3 mmol) of 1-acetoxy-4-acryloxy-2,2,6,6-tetramethylpiperidine and 0.19 g of azobisisobutyronitrile in 30 ml of dry toluene is added over 2 hours to 25 ml of dry toluene maintained at 100°C. The reaction mixture is stirred an additional 15 minutes. A solution of 35 mg of n-dodecyl mercaptan in 2 ml of toluene is added, and the reaction mixture is then poured into cold hexane to yield a precipitate. The precipitate is dissolved in ether, and cold hexane is added to afford a white precipitate which is dried to give 16.4 g (82 % yield) of a brittle glass.
IR: 1755,1720 cm^{-1}.
Anal. Calcd. for $(C_{14}H_{23}NO_4)_n$:
C, 62.4; H, 8.6; N, 5.2.
Found:
C, 62.8; H, 8.6; N, 5.1.

Example 17 1-Acetoxy-2,2,6,6-tetramethylpiperidin-4-yl 4-Hydroxy-3,5-di-tert-butylbenzoate

A solution of 20.0 g (92.9 mmol) of 1-acetoxy-4-hydroxy-2,2,6,6-tetramethylpiperidine in 10.3 g (102 mmol) of triethylamine and 210 ml of dichloromethane is cooled below 0°C. To this solution is added, under nitrogen, a solution of 25.0 g (92.9 mmol) of 4-hydroxy-3,5-di-tert-butylbenzoyl chloride over a 30 minutes interval. During the addition the reaction temperature is maintained at -3 to 0°C. The reaction mixture is stirred at room temperature for 3 hours, then diluted with hexane (200 ml). Triethylamine hydrochloride is removed by filtration, and the filtrate is washed with 1N HCl (200 ml) and saturated sodium bicarbonate solution (200 ml). The solution is dried over magnesium sulfate and concentrated to an oil which is triturated in hexane to yield 1.5 g of a white solid impurity.

The mother liquor is concentrated and crystallized from methanol/dichloromethane to yield 11.6 g (28 %) of a white solid, m.p. 154-156°C, which is the title compound.
Anal. Calcd. for $C_{26}H_{41}NO_5$:
C, 69.7; H, 9.2; N, 3.1.
Found:
C, 69.7; H, 9.4; N, 3.1.

Example 18 Di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) 2-(4-Hydroxy-3,5-di-tert-butylbenzyl) n-butylmalonate

To 10 mmol of lithium diisopropylamide in 25 ml of anhydrous tetrahydrofuran is added a solution of 27.6 g (50 mmol) of di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) n-butylmalonate in 90 ml of THF followed by a solution of 16.5 g (62.5 mmol) of 4-dimethylaminomethyl-2,6-di-tert-butylphenol in 50 ml THF. The reaction mixture is refluxed for 2 hours. The reaction mixture is concentrated under reduced pressure and the residue is dissolved in ether (300 ml). The ether solution is washed with 1N HCl (300 ml) and saturated sodium bicarbonate solution (150 ml), then dried over magnesium sulfate and concentrated. The resulting crude solid is recrystallized from hexane to give 25.7 g (66 % yield) of a white solid, m.p. 174-175°C.
Anal. Calcd. for $C_{44}H_{72}N_2O_8$:
C, 68.4; H, 9.4; N, 3.6.
Found:
C, 68.1; H, 9.4; N, 3.8.

Example 19
N-(1-Acetoxy-2,2,6,6-tetramethylpiperidin-4-yl)-N-(n-butyl)-4-(4-hydroxy-3,5-di-t-butylbenzoyloxy)-3,5-di-t-butylbenzamide

A solution of 20.9 g (77.6 mmol) of 4-hydroxy-3,5-di-tert-butylbenzoyl chloride in 100 ml of dichloromethane is added dropwise over 30 minutes under nitrogen to a chilled solution (0°C) of 20.0 g (64.7 mol) of 1-acetoxy-4-butylamino-2,2,6,6-tetramethylpiperidine hydrochloride in 19.6 g (194 mmol) of triethylamine and 100 ml of dichloromethane. The reaction temperature is maintained below 5°C during the addition. The reaction mixture is stirred for 3 hours at room temperature, then diluted with ether (200 ml) and filtered. The filtrate is washed with 1N HCl (2 x 100 ml) and sodium bicarbonate solution (200 ml). The solution is dried over magnesium sulfate, concentrated, and chromatographed on silica gel (3:1 hexane:ethyl acetate). The major reaction product is crystallized from methanol to yield 13.0 g of a white solid, m.p. 230-232°C.
Anal. Calcd. for $C_{45}H_{70}N_2O_6$:
C, 73.5; H, 9.6; N, 3.8.
Found:
C, 72.6; H, 9.4; N, 3.7.

Example 20 N-(1-Acetoxy-2,2,6,6-tetramethylpiperidin-4-yl)-N-n-butyl-4-hydroxy-3,5-di-t-butylbenzamide
A second recrystallization crop from Example 19 is recrystallized until a constant melting point is obtained. The yield is 4.4 g of a white solid, m.p. 161-164°C, with an analysis consistent with the title compound.
Anal. Calcd. for $C_{30}H_{50}N_2O_4$:
C, 71.7; H, 10.0; N, 5.6.
Found:
C, 72.0; H, 10.2; N, 5.5.

Example 21 1,6-Di-[N-acetyl-N-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl)]-aminohexane
A mixture of 52.8 g (310 mmol) of 1-oxyl-2,2,6,6-tetramethylpiperidin-4-one, 17.4 g (150 mmol) of 1,6-hexanediamine, methanol (10.0 ml), water (150 ml), and platinum oxide (500 mg) is hydrogenated in a Parr apparatus for 18 hours at 50 psi (ambient temperature). Chloroform (1000 ml) is added, and the catalyst is filtered. The organic phase is concentrated to obtain 44.4 g (69 %) of 1,6-di-[N-(1-hydroxy-2,2,6,6-tetramethyl-piperidin-4-yl)]-aminohexane. The hydroxylamine (19.5 g, 45.7 mmol) is added to 150 ml of acetic anhydride over 5 minutes, with the reaction temperature reaching 50°C during the addition. The reaction mixture is refluxed for 30 minutes, then rehydrogenated (400 mg 5 % Pd on C, 50 psi, 3 hours) to reduce any remaining nitroxyl radical. The catalyst is removed by filtration, and solvent is evaporated. The residue is crystallized from 4:1 toluene:heptane to give 14:1 g (52 % yield) of a white solid, m.p. 169-170°C.
Anal. Calcd. for $C_{32}H_{58}N_4O_6$:
C, 64.6; H, 9.8; N, 9.4.
Found:
C, 64.8; H, 9.8; N, 9.3.

Example 22 Di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl) N,N'-(1,6-Hexanediyl)dicarbamate
A mixture of 15.0 g (70 mmol) of 1-acetoxy-4-hydroxy-2,2,6,6-tetramethylpiperidine, 5.9 g (35 mmol) of 1,6-hexanediisocyanate, toluene (125 ml), and dibutyltin dilaurate (200 mg) is refluxed for 1 hour. The hot reaction mixture is poured into hexane (500 ml). The resulting precipitate is removed by filtration and triturated in ether to give 13.1 g (63 % yield) of a white solid, m.p. 158-163°C.
Anal. Calcd. for $C_{30}H_{54}N_4O_6$:
C, 60.2; H, 9.1; N, 9.4.
Found:
C, 60.1; H, 9.1; N, 10.0.

Example 23 1-Acetoxy-4-(N-acetyl-N-n-dodecylamino)-2,2,6,6-tetramethylpiperidine
A mixture of 42.6 g (250 mmol) of 1-oxyl-2,2,6,6-tetramethylpiperidin-4-one, 47.0 g (250 mmol) of n-dodecylamine, 600 mg of platinum oxide and 200 ml of toluene is hydrogenated in a Parr apparatus (50 psi, ambient temperature) for 3 hours. Acetic anhydride (77.9 g, 760 mmol) is added. The catalyst is removed by filtration, and the filtrate is refluxed for 3 hours. Solvent is evaporated at reduced pressure, and the residue is dissolved in 2:1 hexane:ether (500 ml). The solution is washed with 1N HCl (400 ml), saturated sodium bicarbonate solution (200 ml), and saturated sodium chloride solution (200 ml), then dried over magnesium sulfate and concentrated. The residue is chromatographed on silica gel (1:1 hexane:ethyl acetate) to obtain 42.4 g (40 % yield) of a yellow oil.
Anal. Calcd. for $C_{25}H_{48}N_2O_3$:
C, 70.7; H, 11.4; N, 6.6.
Found:
C, 71.0; H, 11.3; N, 6.3.

Example 24 Di-(4-n-octadecanoyloxy-2,2,6,6-tetramethylpiperidin-1-yl) Oxalate

A solution of 4.33 g (34.1 mmol) of oxalyl chloride in 50 ml of dichloromethane is added dropwise over 20 minutes to a solution of 30.0 g (68.2 mmol) of 1-hydroxy-2,2,6,6-tetramethylpiperidin-4-yl) stearate in 8.3 g (81.8 mmol) of triethylamine and 100 ml dichloromethane in a nitrogen atmosphere. The reaction tempeature rises from 20° to 35°C during the addition. The reaction mixture is stirred overnight at room temperature; the triethylamine hydrochloride formed is removed by filtration, and the filtrate is diluted to a total volume of 300 ml with dichloromethane. The solution is washed with 1N HCl (2 x 100 ml) and saturated sodium bicarbonate solution (200 ml), dried over magnesium sulfate, and concentrated to obtain a dark brown solid. The solid is suspended in methanol and filtered. The isolated solid is recrystallized twice from heptane (filtrol added to decolorize) to obtain 9.3 g (29 % yield) of white crystals, m.p. 104-105°C.

Anal. Calcd. for $C_{56}H_{104}N_2O_8$:
C, 72.1; H, 11.2; N, 3.0.
Found:
C, 72.1; H, 11.3; N, 3.2.

Example 25 Di-(4-benzoyloxy-2,2,6,6-tetramethylpiperidin-1-yl) Oxalate

The compound is prepared according to the procedure given for Example 24, utilizing the corresponding benzoate.
m.p. 244°C (dec).
Anal. Calcd. for $C_{34}H_{44}N_2O_8$:
C, 67.1; H, 7.3; N, 4.6.
Found:
C, 66.8; H, 7.4; N, 4.5.

Example 26 Di-(1-benzoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) Sebacate

Benzoyl chloride (29.5 g, 210 mmol) is added dropwise over 30 minutes to a mixture of 51.3 g (100 mmol) of di-(1-hydroxy-2,2,6,6-tetramethylpiperidin-4-yl) sebacate, 250 ml of toluene, and 30.4 g (300 mmol) of triethylamine under nitrogen. The temperature rises from 22° to 48°C during the addition. The reaction mixture is then heated at 70°C for 45 minutes, cooled below 40°C, and diluted with 150 ml of toluene. Triethylamine hydrochloride is filtered off, and the filtrate is washed with 10 % aqueous ammonia (200 ml), warm water (3 x 400 ml), 1N HCl (2 x 200 ml), saturated sodium bicarbonate solution (200 ml) and saturated sodium chloride solution (200 ml). The solution is dried over magnesium sulfate and concentrated. The residue is recrystallized from methanol to obtain 51.5 g (71 % yield) of a white solid, m.p. 92-100°C.

Anal. Calcd. for $C_{42}H_{60}N_2O_8$:
C, 70.0; H, 8.4; N, 3.9.
Found:
C, 69.8; H, 8.6; N, 3,9.

Example 27 1-Benzoyloxy-4-(N-n-butylbenzoylamino)-2,2,6,6-tetramethylpiperidine

The compound is prepared from the reaction of 1-hydroxy-4-(N-n-butylamino-2,2,6,6-tetramethylpiperidine with benzoyl chloride following a procedure similar to that used for Example 26.
m.p. 155-159°C.
Anal. Calcd. for $C_{27}H_{36}N_2O_3$:
C, 74.3; H, 8.3; N, 6.4.
Found:
C, 74.0; H, 8.4; N, 6.3.

Example 28 (1-Benzoyloxy-2,2,6,6-tetramethylpiperidin-4-yl)(1'-Benzyloxy-2,2,6,6-tetramethylpiperidin-4-yl) Isophthalate

A mixture of 35.0 g (78.7 mmol) of di-(2,2,6,6-tetramethylpiperidin-4-yl) isophthalate, 1.0 g of molybdenum hexacarbonyl, and 75 ml of toluene is heated to 90°C in a nitrogen atmosphere. A solution of 4.2 M t-butyl hydroperoxide in toluene (225 ml, 945 mmol) is added over 5 minutes and the reaction mixture turns red. After the addition, the reaction mixture is irradiated for 6 hours (internal temperature 85°C) with a UV lamp. Another 1.0 g portion of molybdenum hexacarbonyl is added, and the reaction mixture is irradiated for 16 hours. The mixture was then filtered and concentrated and the crude residue is chromatographed on silica gel (9:1 hexane:ethyl acetate). The more polar of the two major reaction products is recrystallized from ethanol to obtain 12.0 g (23 % yield) of the title compound, a white solid with m.p. 137-140°C.

Anal. Calcd. for $C_{40}H_{50}N_2O_7$:
C, 71.6; H, 7.5; N, 4.2.
Found:
C, 71.7; H, 7.8; N, 4.4.

Example 29 1,4-Di-(4-hydroxy-3,5-di-tert-butylbenzoyloxy)-2,2,6,6-tetramethylpiperidine

A solution of 31.2 g (116 mmol) of 4-hydroxy-3,5-di-tert-butylbenzoyl chloride in 100 ml of toluene is added dropwise over 3 minutes to a mixture of 10.0 g (58.8 mmol) of 1,4-dihydroxy-2,2,6,6-tetramethylpiperidine and

15.5 g (128 mmol) of N,N-dimethylaniline in 50 ml of toluene in a nitrogen atmosphere. The reaction mixture is heated at 80° for three hours, then diluted to 400 ml. The solution is washed with 1N HCl, saturated sodium bicarbonate solution, and saturated sodium chloride solution, then dried over magnesium sulfate and concentrated. The yellow residue is decolorized with DARCO G-60 in 2-propanol. Crystallization gives 14.0 g (38 % yield) of a white solid, m.p. 196°C(dec).
Anal. Calcd. for $C_{39}H_{59}NO_6$:
C, 73.4; H, 9.3; N, 2.2.
Found:
C, 73.3; H, 9.3; N, 2.1.

Example 30 1-Carbamoyloxy-4-benzoyloxy-2,2,6,6-tetramethylpiperidine
A solution of 6.7 ml of 12 N HCl in 10 ml of water is added dropwise over 10 minutes with cooling below 0°C to a suspension of 20.8 g (75 mmol) of 1-hydroxy-4-benzoyloxy-2,2,6,6-tetramethylpiperidine, 75 ml of methanol, and 25 ml of water in a nitrogen atmosphere. A clear solution resulted. A solution of 6.1 g (75 mmol) of potassium cyanate in 25 ml of water is added dropwise over 30 minutes. The reaction temperature is maintained at 0-5°C during the cyanate addition. The reaction mixture is stirred 30 minutes at ambient temperature, then filtered. The precipitate is washed with water, then dissolved in 200 ml of toluene. Residual water is removed by azeotropic distillation. The toluene solution is then cooled to yield 19.4 g (81 %) of a white crystalline solid, m.p. 148-149°C.
Anal. Calcd. for $C_{17}H_{24}N_2O_4$:
C, 63.7; H, 7.6; N, 8.7.
Found:
C, 63.5; H, 7.8; N, 8.8.

Example 31 Di-(1-carbamoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) Sebacate
The compound is prepared according to the procedure given in Example 30.
Anal. Calcd. for $C_{30}H_{54}N_4O_8$:
C, 60.2; H, 9.1; N, 9,4.
Found:
C, 60.3; H, 9.0; N, 9.1.

Example 32 Di-(1-phenylcarbamoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) Sebacate
Phenyl isocyanate (9.5 g, 80 mmol) is added over 5 minutes to a suspension of 20.0 g (39 mmol) of di-(1-hydroxy-2,2,6,6-tetramethylpiperidin-4-yl) sebacate in 125 ml of dichloromethane. The resulting solution is refluxed for 30 minutes and then evaporated to obtain a solid. The crude solid is briefly refluxed in methanol, filtered off, and recrystallized from 2-propanol:dichloromethane to give 22.2 g (76 % yield) of white solid, m.p. 159-161°C(dec).
Anal. Calcd. for $C_{42}H_{62}N_4O_8$:
C, 67.2; H, 8.3; N, 7.5.
Found:
C, 66.9; H, 8.3; N, 7.3.

Example 33 4-Benzoyloxy-1-n-butylcarbamoyloxy-2,2,6,6-tetramethylpiperidine
A solution of 4.66 g (40 mmol) of n-butyl isocyanate in 10 ml of toluene is added over 10 minutes to a suspension of 10.6 g (38.2 mmol) of 1-hydroxy-4-benzoyloxy-2,2,6,6-tetramethylpiperidine in 40 ml of toluene. The reaction mixture is heated at 50°C for 30 minutes. The reaction mixture is then filtered, and the residue from evaporation of the filtrate is crystallized from hexane to give 4.2 g (29 % yield) of a white solid, m.p. 126-127°C.
Anal. Calcd. for $C_{21}H_{32}N_2O_4$:
C, 67.0; H, 8.6; N, 7.5.
Found:
C, 67.2; H, 8.7; N, 7.4.

Example 34 Di-(1-n-butylcarbamoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) Phthalate
The compound is prepared in 65 % yield according to the procedure given in Example 21, except that the appropriate phthalate and dichloromethane solvent are utilized.
m.p. 180-182°C.
Anal. Calcd. for $C_{36}H_{58}N_4O_8$:
C, 64.1; H, 8.7; N, 8.3.
Found:
C, 63.9; H, 8.8; N, 8.0.

Example 35 Di-(1-n-butylcarbamoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) Isophthalate
The compound is prepared according to the procedure given in Example 34 except for the use of the isophthalate.

m.p. 176-177°C.
Anal. Calcd. for $C_{36}H_{58}N_4O_8$:
C, 64.1; H, 8.7; N, 8.3.
Found:
C, 64.1; H, 8.4; N, 8.3.

Example 36 Di-(1-n-butylcarbamoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) 2,2-Diethylmalonate
The compound is prepared according to the procedure given in Example 34 except for the use of the appropriate malonate.
m.p. 166-169°C.
Anal. Calcd. for $C_{35}H_{64}N_4O_8$:
C, 62.8; H, 9.6; N, 8.4.
Found:
C, 63.2; H, 9.6; N, 8.3.

Example 37 Di-(1-n-butylcarbamoyloxy-2,2,6,6-tetramethylpiperidin-4-yl) n-Butylmalonate
The compound is prepared according to the procedure given in Example 34. m.p. 129-131°C.
Anal. Calcd. for $C_{35}H_{64}N_4O_8$:
C, 62.8; H, 9.6; N, 8.4.
Found:
C, 63.1; H, 9.5; N, 8.3.

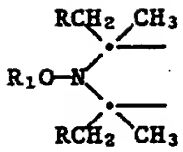
Example 38 Di-(4-benzoyloxy-2,2,6,6-tetramethylpiperidin-1-yl)
N,N'-(2,4,4-trimethyl-1,6-hexanediyl)-dicarbamate
The compound is prepared from 4-benzoyloxy-1-hydroxy-2,2,6,6-tetramethylpiperidine and 2,4,4-trimethylhexane-1,6-diisocyanate according to the procedure given in Example 33.
m.p. 165-167°C.
Anal. Calcd. for $C_{43}H_{64}N_4O_8$:
C, 67.5; H, 8.4; N, 7.3.
Found:
C, 67,4; H, 9.0; N, 7.4.

Example 39 n-Butyl (4-Benzoyloxy-2,2,6,6-tetramethylpiperidin-1-yl) Carbonate
A solution of 7.8 g (81 mmol) of n-butylchloroformate in 15 ml of toluene is added dropwise over 20 minutes to a mixture of 15.0 g (54 mmol) of 1-hydroxy-4-benzoyloxy-2,2,6,6-tetramethylpiperidine, 8.2 ml (101 mmol) of triethylamine, and 75 ml of toluene in a nitrogen atmosphere. During the addition, the reaction temperature is maintained below 10°C with an ice bath. After the addition is complete, the reaction mixture is stirred two hours at room temperature. Triethylamine hydrochloride is filtered, the filtrate washed with 1N HCl (200 ml), saturated sodium bicarbonate (200 ml), and saturated sodium chloride (200 ml), then dried over magnesium sulfate and concentrated. The resulting oil is crystallized from hexane to give 14.1 g (69 % yield) of a white solid, m.p. 83-84°C.
Anal. Calcd. for $C_{21}H_{31}NO_4$:
C, 66.8; H, 8.3; N, 3.7.
Found:
C, 66.8; H, 8.4; N, 3.9.

Summarizing, this invention is seen to provide a series of new O-acyl, O-carbamoyl and O-carbonate substituted N-hydroxy hindered amine stabilizers. Variations may be made in proportions, procedures and materials without departing from the scope of the invention as defined by the following claims.

Claims

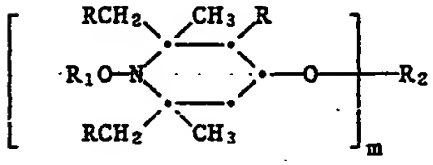
1. A stabilized ambient curable or acid catalyzed thermosetting coating composition containing an effective stabilizing amount of a hindered amine compound containing the group



wherein R is hydrogen or methyl and R_1 is a group

D-C(=O)-, wherein D is C_1-C_{18} alkyl, C_1-C_{18} alkoxy, phenyl, phenyl substituted by hydroxy, C_1-C_4 alkyl or C_1-C_4 alkoxy, or D is amino or amino mono- or disubstituted by C_1-C_{12} alkyl or phenyl.

2. The composition according to claim 1, which contains a hindered amine compound corresponding to one of the formulae A-N



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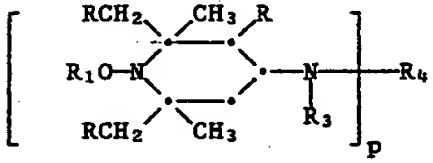

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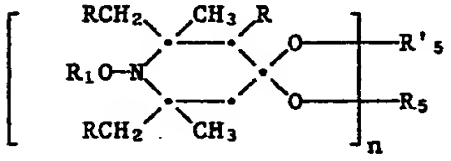

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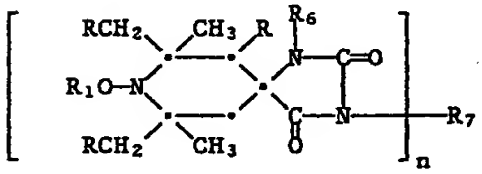

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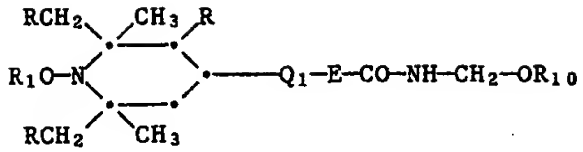

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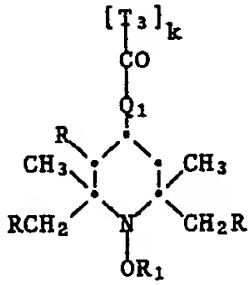

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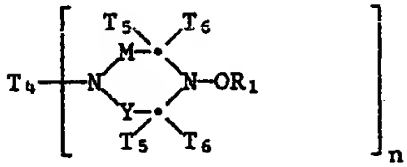

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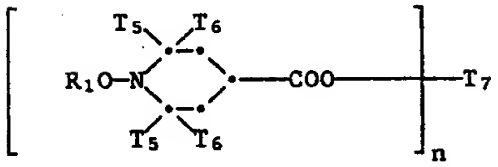
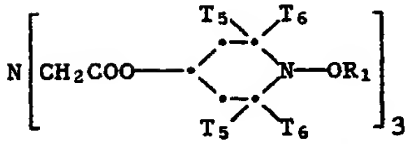


(H)



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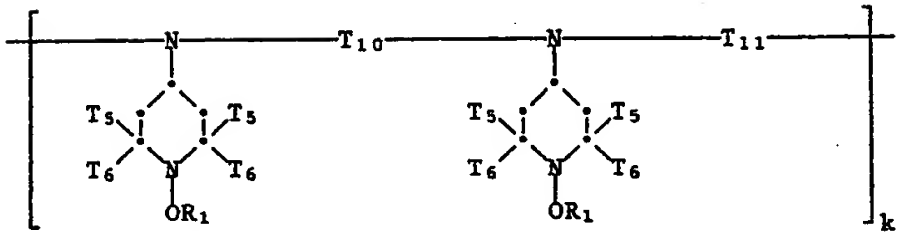
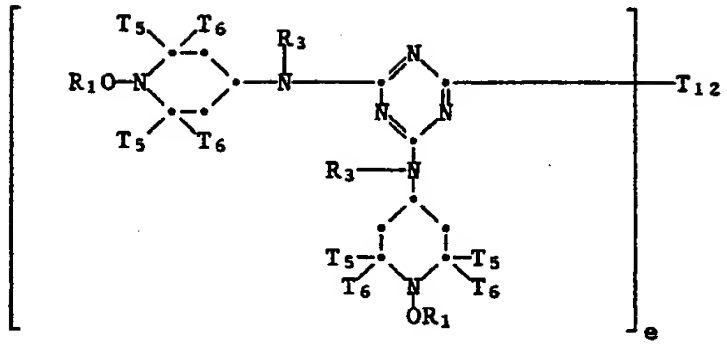
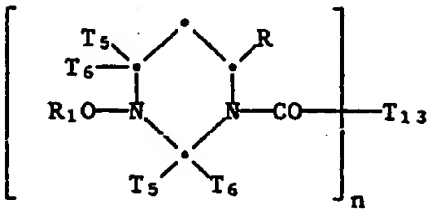


(J)



(K)



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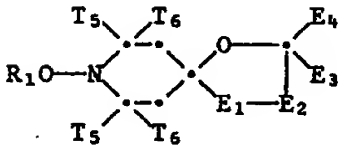

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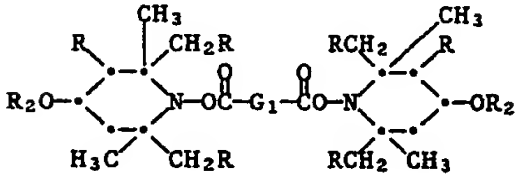
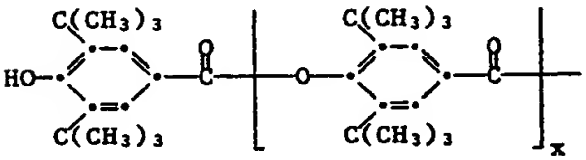
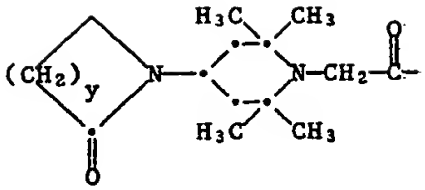


(N)

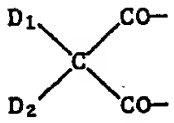
wherein
R is hydrogen or methyl,
R^1 is a group D-CO- , wherein D is C_1-C_{18} alkyl, C_1-C_{18} alkoxy, phenyl, phenyl substituted by hydroxy, C_1-C_4 alkyl or C_1-C_4 alkoxy, or amino or amino mono- or disubstituted by C_1-C_{12} alkyl or phenyl;
m is 1-4,
when m is 1,
R_2 is hydrogen, C_1-C_{18} alkyl optionally interrupted by one or more oxygen atoms, C_2-C_{12} alkenyl, C_6-C_{10} aryl, C_7-C_{18} aralkyl, glycidyl, a monovalent acyl radical of an aliphatic, cycloaliphatic, araliphatic or aromatic carboxylic acid, or of a carbamic acid, or R_2 is a group



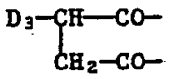
wherein x is 0 or 1, or is a group



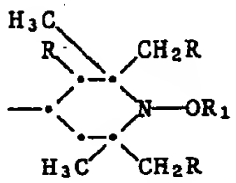
wherein y is 2-4;
when m is 2,
R_2 is C_1-C_{12} alkylene, C_4-C_{12} alkenylene, xylylene, a divalent acyl radical of an aliphatic, cycloaliphatic, araliphatic or aromatic dicarboxylic acid or of a dicarbamic acid, or R_2 is a group



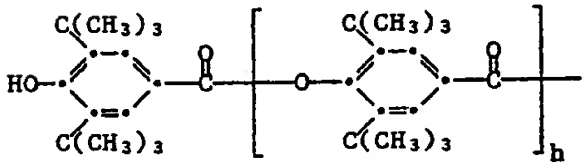
or



wherein D_1 and D_2 independently are hydrogen, an alkyl radical containing up to 8 carbon atoms, phenyl, benzyl or 3,5-di-t-butyl-4-hydroxybenzyl, and D_3 is an alkyl or alkenyl radical containing up to 18 carbon atoms;
when m is 3, R_2 is a trivalent acyl radical of an aliphatic, cycloaliphatic, or aromatic tricarboxylic acid; when m is 4, R_2 is a tetravalent acyl radical of a saturated or unsaturated aliphatic or aromatic tetracarboxylic acid; p is 1, 2 or 3,
R_3 is hydrogen, C_1-C_{12} alkyl, C_5-C_7 cycloalkyl, C_7-C_9 aralkyl, C_2-C_{18} alkanoyl, C_3-C_5 alkenoyl or benzoyl;
when p is 1,
R_4 is hydrogen, C_1-C_{18} alkyl, C_5-C_7 cycloalkyl, C_2-C_8 alkenyl unsubstituted or substituted by a cyano, carbonyl or carbamide group, or it is aryl, aralkyl, glycidyl, a group of the formula $-CH_2-CH(OH)-Z$ or of the formula -CONH-Z wherein Z is hydrogen, methyl or phenyl;
or R_4 is a group of the formula I

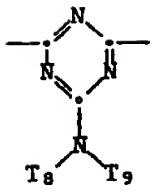


(I) or



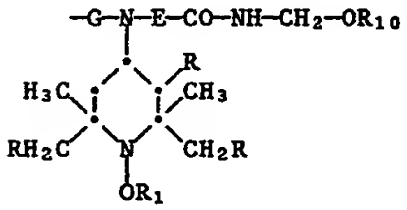
with h as 0 or 1;

or R_3 and R_4 together are alkylene of 4 to 6 carbon atoms or 1-oxo alkylene or the divalent acyl radical of an aliphatic or aromatic 1,2- or 1,3-dicarboxylic acid;
when p is 2;
R_4 is C_1-C_{12} alkylene, C_6-C_{12} arylene, xylylene, a $-CH_2CH(OH)-CH_2$ group, or a group $-CH_2-CH(OH)-CH_2-O-X-O-CH_2-CH(OH)-CH_2-$ wherein X is C_2-C_{10} alkylene, C_6-C_{15} arylene or C_6-C_{12} cycloalkylene; or, provided that R_3 is not alkanoyl, alkenoyl or benzoyl, R_4 can also be a divalent acyl radical of an aliphatic, cycloaliphatic or aromatic dicarboxylic or dicarbamic acid, or can be the group -CO-; or R_4 is a group of formula II



(II)

where T_8 and T_9 are independently hydrogen, alkyl of 1 to 18 carbon atoms, or T_8 and T_9 together are alkylene of 4 to 6 carbon atoms or 3-oxapentamethylene, preferably T_8 and T_9 together are 3-oxapentamethylene;
when p is 3, R_4 is 2,4,6-triazinetriyl;
n is 1 or 2 and
when n is 1,
R_5 and R'_5 are independently C_1-C_{12} alkyl, C_2-C_{12} alkenyl, C_7-C_{12} aralkyl, or R_5 is also hydrogen, or R_5 and R'_5 together are C_2-C_8 alkylene or hydroxyalkylene or C_4-C_{22} acyloxyalkylene;
when n is 2,
R_5 and R'_5 together are $(-CH_2)_2C(CH_2-)_2$;
R_6 is hydrogen, C_1-C_{12} alkyl, allyl, benzyl, glycidyl or C_2-C_6 alkoxyalkyl;
when n is 1,
R_7 is hydrogen, C_1-C_{12} alkyl, C_3-C_5 alkenyl, C_7-C_9 aralkyl, C_5-C_7 cycloalkyl, C_2-C_4 hydroxyalkyl, C_2-C_6 alkoxyalkyl, C_6-C_{10} aryl, glycidyl, a group of the formula $-(CH_2)_t-COO-Q$ or of the formula $-(CH_2)_t-O-CO-Q$ wherein t is 1 or 2, and Q is C_1-C_4 alkyl or phenyl; or
when n is 2,
R_7 is C_2-C_{12} alkylene, C_6-C_{12} arylene, a group $-CH_2CH(OH)-CH_2-O-X-O-CH_2-CH(OH)-CH_2-$ wherein X is C_2-C_{10} alkylene, C_6-C_{15} arylene or C_6-C_{12} cycloalkylene, or a group $-CH_2CH(OZ')CH_2-(OCH_2-CH(OZ')CH_2)_2-$ wherein Z' is hydrogen, C_1-C_{18} alkyl, allyl, benzyl, C_2-C_{12} alkanoyl or benzoyl;
Q_1 is $-N(R_8)-$ or $-O-$;
E is C_1-C_3 alkylene, the group $-CH_2-CH(R_9)-O-$ wherein R_9 is hydrogen, methyl or phenyl, or E is the group $-(CH_2)_3-NH-$ or a direct bond;
R_{10} is hydrogen or C_1-C_{18} alkyl;
R_8 is hydrogen, C_1-C_{18} alkyl, C_5-C_7 cycloalkyl, C_7-C_{12} aralkyl, cyanoethyl, C_6-C_{10} aryl, the group $-CH_2-CH(R_9)-OH$ wherein R_9 has the meaning defined above; a group of the formula I
or a group of the formula



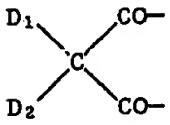
wherein G can be C_2-C_8 alkylene or C_6-C_{12} arylene; or R_8 is a group $-E-CO-NH-CH_2-OR_{10}$;
T_3 is ethylene or 1,2-propylene, or is the repeating structural unit derived from an alpha-olefin copolymer with an alkyl acrylate or methacrylate;
k is 2 to 100;
T_4 has the same meaning as R_4 when p is 1 or 2,
T_5 is methyl,
T_6 is methyl or ethyl, or T_5 and T_6 together are tetramethylene or pentamethylene;
M and Y are independently methylene or carbonyl;
T_7 is the same as R_7;
T_{10} and T_{11} are independently alkylene of 2 to 12 carbon atoms, or T_{11} is a group of formula II;
e is 2, 3 or 4 and
T_{12} is a group $-N(R_5)-(CH_2)_d-N(R_5)-$ or

$-NH(CH_2)_a-\overset{\downarrow}{N}(CH_2)_b-\overset{\downarrow}{N}[(CH_2)_c-\overset{\downarrow}{N}]_fH$ where a, b and c are independently 2 or 3, d is 2 to 10 and f is 0 or 1;
T_{13} is the same as R_4 with the proviso that R_{13} cannot be hydrogen when n is 1;
E_1 and E_2, being different, each are -CO- or $-N(E_5)-$, where E_5 is hydrogen, C_1-C_{12} alkyl or C_4-C_{22}

alkoxycarbonylalkyl;
E_3 is hydrogen, alkyl of 1 to 30 carbon atoms, phenyl, naphthyl, said phenyl or said naphthyl substituted by chlorine or by alkyl of 1 to 4 carbon atoms, or phenylalkyl of 7 to 12 carbon atoms, or said phenylalkyl substituted by alkyl of 1 to 4 carbon atoms,
E_4 is hydrogen, alkyl of 1 to 30 carbon atoms, phenyl, naphthyl or phenylalkyl of 7 to 12 carbon atoms, or E_3 and E_4 together are polymethylene of 4 to 17 carbon atoms, or said polymethylene substituted by up to four alkyl groups of 1 to 4 carbon atoms;
R_2 of formula (N) is as previously defined when m is 1;
G_1 is a direct bond, C_1-C_{12} alkylene, phenylene or -NH-G'-NH wherein G' is C_1-C_{12} alkylene.

3. A composition according to claim 2 which contains a compound of formulae A, B, D, J, K or M wherein R is hydrogen and T_5 and T_6 are methyl.

4. A composition according to claim 2 which contains a compound of formula A wherein R is hydrogen, R_1 is a group D-CO- and D is C_1-C_{12} alkyl, C_1-C_4 alkoxy, phenyl, amino or amino monosubstituted by C_1-C_{12} alkyl or phenyl,
m is 1, 2 or 4 and when m is 1,
R_2 is an acyl radical of an aliphatic C_2-C_{18} carboxylic acid, of a cycloaliphatic C_6-C_{12} carboxylic acid or of an aromatic C_7-C_{15} carboxylic acid and when m is 2, R_2 is a divalent acyl radical of an aliphatic C_2-C_{18} dicarboxylic acid or of a cycloaliphatic or aromatic C_8-C_{14} dicarboxylic acid, or of an aliphatic, cycloaliphatic or aromatic C_8-C_{14} dicarbamic acid, or R_2 is a group



wherein D_1 is C_1-C_8 alkyl or 3,5-di-tert.butyl-4-hydroxybenzyl and D_2 is D_1 or hydrogen and when m is 4, R_2 is a tetravalent acyl radical of a butane- or pentane-tetracarboxylic acid.

5. A composition according to claim 2 which contains a compound of formula B wherein R is hydrogen, R_1 is a group D-CO- and D is C_1-C_{12} alkyl, C_1-C_4 alkoxy, phenyl, amino or amino monosubstituted by C_1-C_{12} alkyl or phenyl,
p is 1 or 2, R_3 is hydrogen, C_1-C_{12} alkyl or C_2-C_{12} alkanoyl and when p is 1, R_4 is C_1-C_{12} alkyl, C_5-C_7 cycloalkyl or a group of formula I,
and when p is 2, R_4 is C_2-C_8 alkylene, phenylene or xylylene, and if R_3 is not alkanoyl, R_4 may also be a divalent acyl residue of an aliphatic C_2-C_{10} dicarboxylic acid or of an aromatic C_8 dicarboxylic acid or of an aliphatic or aromatic C_8-C_{15} dicarbamic acid.

6. A composition of claim 1, which contains di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl)sebacate.

7. The composition of claim 3, which contains alpha,alpha'-(di-1-ethoxy-2,2,6,6-tetramethylpiperidin-4-yloxy)-p-xylene.

8. The composition of claim 3, containing 1,4-dibenzyloxy-2,2,6,6-tetramethylpiperidine.

9. The composition of claim 3, containing di-(1-benzyloxy-2,2,6,6-tetramethylpiperidin-4-yl) sebacate.

The composition of claim 3, containing 4-benzyloxy-1-ethoxy-2,2,6,6-tetramethylpiperidine.

11. The composition of claim 3, containing 1,4-di-(4-hydroxy-3,5-di-tert-butylbenzoyloxy)-2,2,6,6-tetramethylpiperidine.

12. The composition of claim 3, containing alpha,alpha'-(di-1-benzoyloxy-2,2,6,6-tetramethylpiperidin-4-yloxy)-p-xylene.

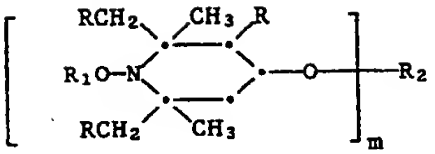
13. A coating composition according to claim 1, which is an ambient curable system based on an alkyd resin, thermoplastic acrylic resin, acrylic alkyd resin, polyurethane resin or polyester resin, or said resins modified with silicones, isocyanates, epoxides, isocyanurates, ketimines or oxazolidines, or the system is based on a cellulose ester or on an epoxide resin.

14. A coating composition according to claim 1, which is an acid catalyzed thermosetting system based on a hot crosslinkable acrylic, polyester, polyurethane, polyamide or alkyd resin.

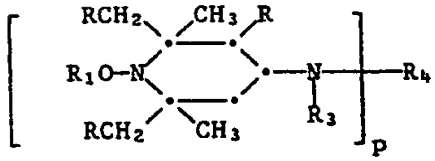
15. The composition according to claim 1, which is an enamel for industrial finishes.

16. The composition according to claim 1, which is a refinishing enamel for automobiles.

17. A compound corresponding to one of the formulae A' to N'



(A')



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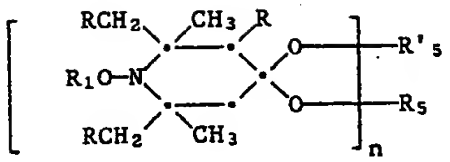

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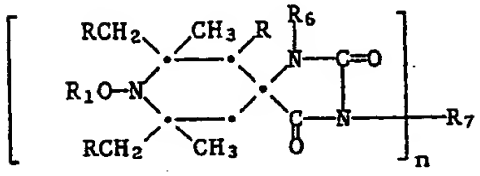

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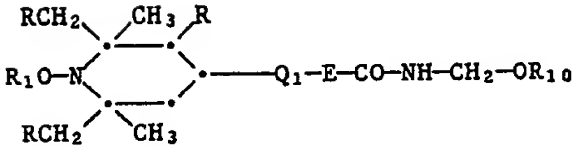

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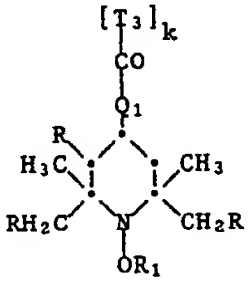

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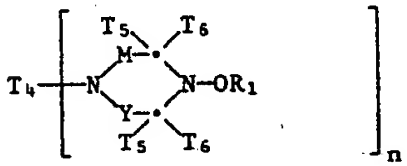

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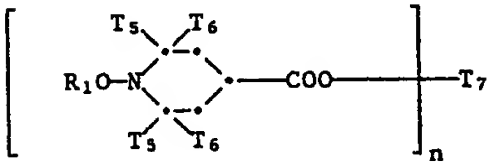

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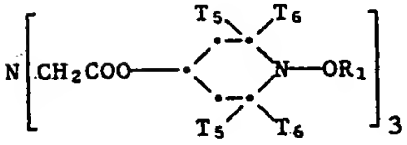

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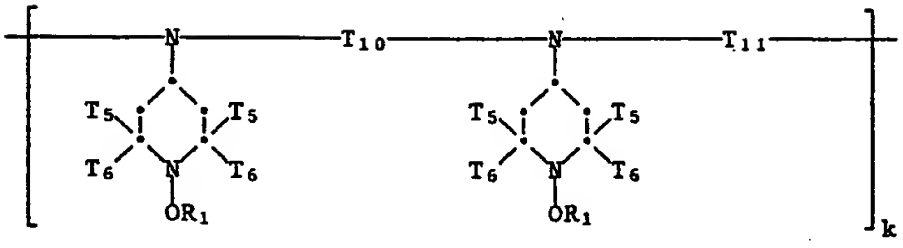

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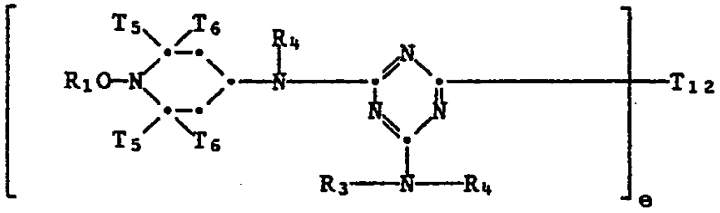

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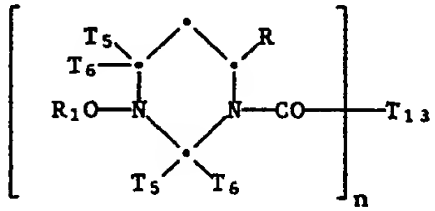

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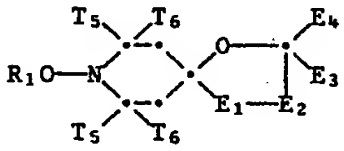

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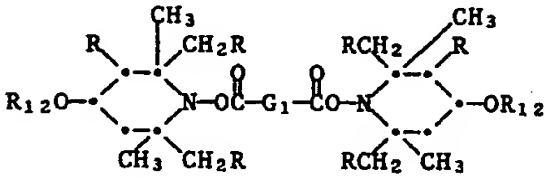
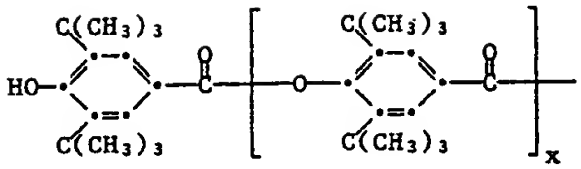


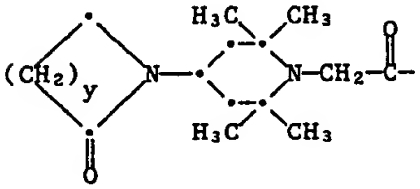
(N')

wherein
R is hydrogen or methyl,
R_1 is a group

wherein D is C_1-C_{18} alkyl, C_1-C_{18} alkoxy, phenyl, phenyl substituted by hydroxy, C_1-C_4 alkyl or C_1-C_4 alkoxy, or amino or amino mono- or disubstituted by C_1-C_{12} alkyl or phenyl;
m is 1-4,
when m is 1
R_2 is a group



wherein x is 0 or 1, or R_2 is a group

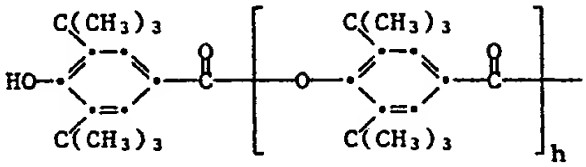


wherein y is 2-4;
when m is 2 and D is alkyl,
R_2 is C_1-C_{12} alkylene, C_4-C_{12} alkenylene, xylylene, a divalent acyl radical of a cycloaliphatic, araliphatic or aromatic dicarboxylic acid having 8-14 C atoms or of an aliphatic, cycloaliphatic or aromatic dicarbamic acid having 8-14 C atoms, or R_2 is a group

wherein D_1 is C_1-C_8 alkyl, benzyl or 3,5-di-t-butyl-4-hydroxybenzyl, D_2 is D_1 or hydrogen, D_3 is C_1-C_{18} alkyl or C_2-C_{18} alkenyl;
when m is 2 and D is phenyl or substituted phenyl, amino, substituted amino or alkoxy,
R_2 is C_1-C_{12} alkylene, C_4-C_{12} alkenylene, xylylene, a divalent acyl radical of an aliphatic, cycloaliphatic, araliphatic or aromatic dicarboxylic or dicarbamic acid having up to 14 C atoms; or R_2 is a group

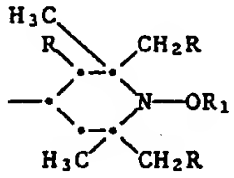
wherein D_4 and D_5 are independently hydrogen, C_1-C_8 alkyl, benzyl or 3,5-di-t-butyl-4-hydroxybenzyl and D_6 is C_1-C_{18} alkyl or C_2-C_{18} alkenyl;
when m is 3, R_2 is a trivalent acyl radical of an aliphatic, cycloaliphatic, or aromatic tricarboxylic acid having up to 12 C atoms;
when m is 4, R_2 is a tetravalent acyl radical of a saturated or unsaturated aliphatic or aromatic tetracarboxylic acid having up to 18 C atoms;
p is 1, 2 or 3,
R_3 is hydrogen, C_1-C_{12} alkyl, C_5-C_7 cycloalkyl, C_7-C_9 aralkyl, C_2-C_{18} alkanoyl, C_3-C_5 alkenoyl or benzoyl;
when p is 1,
R_4 is hydrogen, C_1-C_{18} alkyl, C_5-C_7 cycloalkyl, C_2-C_8 alkenyl unsubstituted or substituted by a cyano, carbonyl or carbamide group, or it is aryl, aralkyl, glycidyl, a group of the formula $-CH_2-CH(OH)-Z$ or

-CONH-Z wherein Z is hydrogen, methyl or phenyl;
or R_4 is a group of the formula



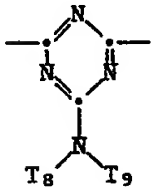
with h as 0 or 1;

or a group of formula I



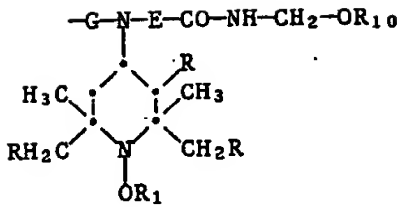
(I);

or R_3 and R_4 together are alkylene of 4 to 6 carbon atoms or 1-oxo alkylene or the divalent acyl radical of an aliphatic or aromatic 1,2- or 1,3-dicarboxylic acid;
when p is 2,
R_4 is C_1-C_{12} alkylene, C_6-C_{12} arylene, xylylene, a $-CH_2CH(OH)-CH_2-$ group, or a group $-CH_2-CH(OH)-CH_2-O-X-O-CH_2-CH(OH)-CH_2-$ wherein X is C_2-C_{10} alkylene, C_6-C_{15} arylene or C_6-C_{12} cycloalkylene; or, provided that R_3 is not alkanoyl, alkenoyl or benzoyl, R_4 can also be a divalent acyl radical of an aliphatic, cycloaliphatic or aromatic dicarboxylic or dicarbamic acid having up to 14 C atoms, or can be the group -CO- or a group of formula II

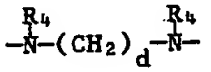


(II)

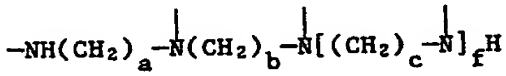
where T_8 and T_9 are independently hydrogen, alkyl of 1 to 18 carbon atoms, or T_8 and T_9 together are alkylene of 4 to 6 carbon atoms or 3-oxapentamethylene;
when p is 3, R_4 is 2,4,6-triazinetriyl,
n is 1 or 2, and when n is 1,
R_5 and R'_5 are independently C_1-C_{12} alkyl, C_2-C_{12} alkenyl, C_7-C_{12} aralkyl, or R_5 is also hydrogen, or R_5 and R'_5 together are C_2-C_8 alkylene or hydroxyalkylene or C_4-C_{22} acyloxyalkylene;
and when n is 2,
R_5 and R'_5 together are $(-CH_2)_2C(CH_2-)_2$;
R_6 is hydrogen, C_1-C_{12} alkyl, allyl, benzyl, glycidyl or C_2-C_6 alkoxyalkyl;
when n is 1,
R_7 is hydrogen, C_1-C_{12} alkyl, C_3-C_5 alkenyl, C_7-C_9 aralkyl, C_5-C_7 cycloalkyl, C_2-C_4 hydroxyalkyl, C_2-C_6 alkoxyalkyl, C_6-C_{10} aryl, glycidyl, a group of the formula $-(CH_2)_t-COO-Q$ or of the formula $-(CH_2)_t-O-CO-Q$ wherein t is 1 or 2, and Q is C_1-C_4 alkyl or phenyl; and
when n is 2,
R_7 is C_2-C_{12} alkylene, C_6-C_{12} arylene, a group $-CH_2CH(OH)-CH_2-O-X-O-CH_2-CH(OH)-CH_2-$ wherein X is C_2-C_{10} alkylene, C_6-C_{15} arylene or C_6-C_{12} cycloalkylene, or a group $-CH_2CH(OZ')CH_2-(OCH_2-CH(OZ')CH_2)_2-$ wherein Z' is hydrogen, C_1-C_{18} alkyl, allyl, benzyl, C_2-C_{12} alkanoyl or benzoyl;
Q_1 is $-N(R_8)-$ or $-O-$; E is C_1-C_3 alkylene, the group $-CH_2-CH(R_9)-O-$ wherein R_9 is hydrogen, methyl or phenyl, or E is the group $-(CH_2)_3-NH-$ or a direct bond;
R_{10} is hydrogen or C_1-C_{18} alkyl, R_8 is hydrogen, C_1-C_{18} alkyl, C_5-C_7 cycloalkyl, C_7-C_{12} aralkyl, cyanoethyl, C_6-C_{10} aryl, the group $-CH_2-CH(R_9)-OH$ wherein R_9 has the meaning defined above; a group of the formula I or a group of the formula



wherein G can be C_2-C_6 alkylene or C_6-C_{12} arylene; or R_8 is a group -E-CO-NH-CH_2-OR_{10};
T_3 is ethylene or 1,2-propylene, or is the repeating structural unit derived from an alpha-olefin copolymer with an alkyl acrylate or methacrylate;
k is 2 to 100;
T_4 has the same meaning as R_4 when p is 1 or 2,
T_5 is methyl,
T_6 is methyl or ethyl, or T_5 and T_6 together are tetramethylene or pentamethylene,
M and Y are independently methylene or carbonyl;
T_7 is the same as R_7,
T_{10} and T_{11} are independently alkylene of 2 to 12 carbon atoms, or T_{11} is a group of formula II;
e is 2, 3 or 4,
T_{12} is a group



or



where a, b and c are independently 2 or 3, d is 2-10 and f is 0 or 1;
T_{13} is the same as R_4 with the proviso that T_{13} cannot be hydrogen when n is 1;
E_1 and E_2, being different, each are -CO- or -N(E_5)- where E_5 is hydrogen, C_1-C_{12} alkyl or alkoxycarbonylalkyl of 4 to 22 carbon atoms;
E_3 is hydrogen, alkyl of 1 to 3 carbon atoms, phenyl, naphthyl, said phenyl or said naphthyl substituted by chlorine or by alkyl of 1 to 4 carbon atoms, or phenylalkyl of 7 to 12 carbon atoms, or said phenylalkyl substituted by alkyl of 1 to 4 carbon atoms,
E_4 is hydrogen, alkyl of 1 to 30 carbon atoms, phenyl, naphthyl or phenylalkyl of 7 to 12 carbon atoms, or E_3 and E_4 together are polymethylene of 4 to 17 carbon atoms, or said polymethylene substituted by up to four alkyl groups of 1 to 4 carbon atoms; and
R_{12} is C_2-C_{18} alkanoyl, C_3-C_6 alkenoyl, benzoyl or benzoyl substituted by C_1-C_4 alkyl, halogen or hydroxyl, and G_1 is a direct bond, C_1-C_{12} alkylene, phenylene or -NH-G'-NH wherein G' is C_1-C_{12} alkylene.

18. A compound of claim 17, wherein D is C_1-C_{18} alkyl, phenyl or phenyl substituted by hydroxy, C_1-C_4 alkyl or C_1-C_4 alkoxy.

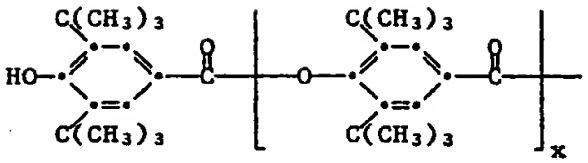
19. A compound of claim 17, wherein D is amino or amino mono- or di-substituted by C_1-C_{12} alkyl or phenyl.

20. A compound of claim 17, wherein D is C_1-C_{18} alkoxy.

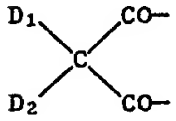
21. A compound of claim 17, wherein R is hydrogen.

22. A compound of claim 17, wherein R_1 is -CO-D and D is C_1-C_{12} alkyl, C_1-C_4 alkoxy, phenyl, amino, C_1-C_{12} alkylamino or phenylamino.

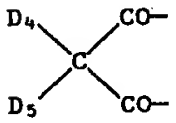
23. A compound of claim 17 of formula A', B', D', F', J', K', M' or N', wherein R is hydrogen,
R_1 is defined in claim 22,
m is 1, 2 or 4, and
when m is 1, R_2 is



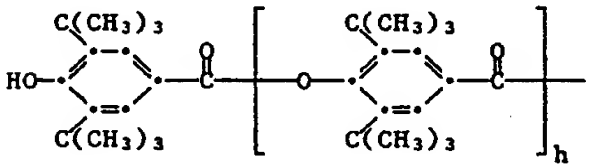
wherein x is 0 or 1,
when m is 2 and D is alkyl,
R_2 is a divalent acyl radical of a cycloaliphatic or aromatic dicarboxylic acid having 8-14 C atoms, or of an aliphatic, cycloaliphatic or aromatic dicarbamic acid having 8-14 C atoms; or R_2 is a group



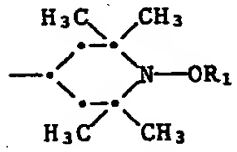
wherein D_1 is C_1-C_4 alkyl or 3,5-di-t-butyl-4-hydroxybenzyl and D_2 is D_1 or hydrogen, and when m is 2 and D is phenyl, amino, alkylamino, phenylamino or alkoxy, R_2 is a divalent acyl radical of an aliphatic, dicarboxylic acid having 2-8 C atoms or of a cycloaliphatic or aromatic dicarboxylic acid having 8-14 C atoms or of an aliphatic, cycloaliphatic or aromatic dicarbamic acid having 8-14 C atoms, or R_2 is a group



wherein D_4 and D_5 are independently hydrogen, C_1-C_8 alkyl, benzyl or 3,5-di-t-butyl-4-hydroxybenzyl, and when m is 4, R_2 is a tetravelent acyl radical of 1,2,3,4-butanetetracarboxylic acid, 1,2,3,4-but-2-enetetracarboxylic acid, 1,2,3,5- or 1,2,4,5-pentanetetracarboxylic acid;
p is 1, 2 or 3,
R_3 is hydrogen, C_1-C_{12} alkyl, cyclohexyl, C_7-C_9 aralkyl, C_2-C_{18} alkanoyl or benzoyl;
when p is 1,
R_4 is C_1-C_{18} alkyl, cyclohexyl, allyl, benzyl or a group

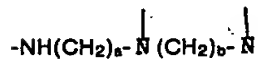


with h as 0 or 1, or is a group



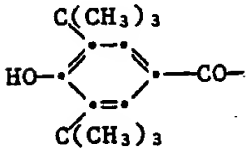
when p is 2,

R_4 is C_1-C_{12} alkylene, C_6-C_{12} arylene, xylylene, a group $-CH_2CH(OH)-CH_2-$, or provided that R_3 is not alkanoyl or benzoyl, R_4 can also be a divalent acyl radical of an aliphatic, cycloaliphatic or aromatic dicarboxylic acid or dicarbamic acid having 6-12 C atoms,
or R_4 is a group of formula II,
where T_8 and T_9 are independently hydrogen or C_1-C_{12} alkyl or T_8 and T_9 together are C_4-C_6 alkylene or 3-oxapentamethylene,
when p is 3, R_4 is 2,4,6-triazinetriyl;
n is 1 or 2 and
R_6 is hydrogen, C_1-C_{12} alkyl, allyl or benzyl,
when n is 1, R_7 is hydrogen, C_1-C_{12} alkyl, allyl, benzyl or cyclohexyl,
and when n is 2, R_7 is C_2-C_{12} alkylene;
k is 5 to 50, T_3 is ethylene or propylene and Q_1 is NH or O;
T_5 and T_6 each are methyl,
T_{10} and T_{11} are independently C_2-C_{12} alkylene or T_{11} is a group of formula II,
e is 4 and
T_{12} is

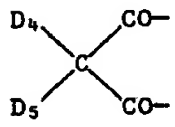


where a, b and c are independently 2 or 3;
E_1 is -CO- and E_2 is -N(E_5)-, E_3 and E_4 independently are C_1-C_{12} alkyl or E_3 and E_4 together are C_5-C_{12} polymethylene, and E_5 is hydrogen or C_1-C_{12} alkyl;
G_1 is a direct bond, C_2-C_8 alkylene or -NH-G'-NH- wherein G' is C_2-C_8 alkylene, and R_{12} is C_2-C_{18} alkanoyl or benzoyl.

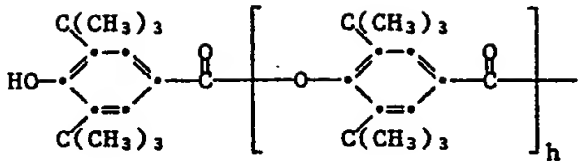
24. A compound of claim 17 of formula A', B', F', J' or N', wherein R is hydrogen,
R_1 is -CO-D and D is C_1-C_4 alkyl, C_1-C_4 alkoxy, phenyl, amino, C_1-C_4 alkylamino or phenylamino;
m is 1 or 2 and
when m is 1, R_2 is a group



when m is 2 and D is alkyl, R_2 is a divalent acyl radical of a phenylenedicarboxylic acid or is a group $-CO-C(D_1)(D_2)-CO-$ wherein D_1 is C_1-C_4 alkyl or 3,5-di-t-butyl-4-hydroxybenzyl and D_2 is D_1 or hydrogen, and when m is 2 and D is alkoxy, phenyl, amino or alkylamino,
R_2 is a divalent acyl radical of an aliphatic or aromatic dicarboxylic acid having 8-10 C atoms or of an aliphatic dicarbamic acid having 8-10 C atoms; or R_2 is a group



wherein D_4 is alkyl or 3,5-di-t-butyl-4-hydroxybenzyl and D_5 is D_4 or hydrogen;
p is 1 or 2, R_3 is C_1-C_4 alkyl, acetyl or benzoyl,
when p is 1, R_4 is C_1-C_{12} alkyl or a group

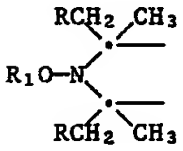


with h as 0 or 1,
and when p is 2, R_4 is C_4-C_8 alkylene;
k is 5 to 50, T_3 is ethylene and Q_1 is 0;
T_5 and T_6 are methyl,
T_{10} is hexamethylene and T_{11} is a group of formula II
wherein T_8 and T_9 are independently hydrogen or C_1-C_{12} alkyl or T_8 and T_9 together are pentamethylene or 3-oxapentamethylene;
R_{12} is acetyl or benzoyl and G_1 is a direct bond or -NH-$(CH_2)_6$-NH-.

25. Use of a compound of claim 17 as stabilizer for organic materials against actinic degradation.
26. Use according to claim 25, as stabilizer for organic polymers.
27. Use according to claim 25, as stabilizer for photographic layers.
28. Organic material containing an effective stabilizing amount of a compound of claim 17.

Claims for the following Contracting State: ES

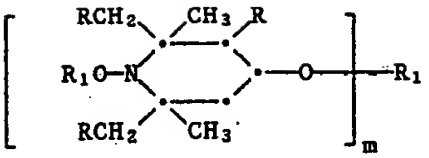
1. A stabilized ambient curable or acid catalyzed thermosetting coating composition containing an effective stabilizing amount of a hindered amine compound containing the group



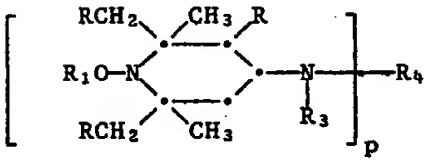
wherein R is hydrogen or methyl and R_1 is a group

D-C(=O)- , wherein D is C_1-C_{18} alkyl, C_1-C_{18} alkoxy, phenyl, phenyl substituted by hydroxy, C_1-C_4 alkyl or C_1-C_4 alkoxy, or D is amino or amino mono- or disubstituted by C_1-C_{12} alkyl or phenyl.

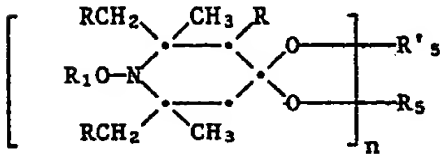
2. The composition according to claim 1, which contains a hindered amine compound corresponding to one of the formulae A-N



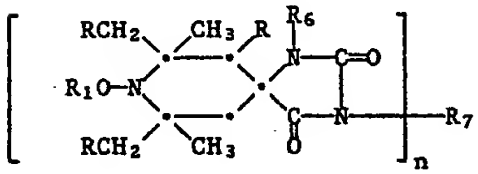
(A)



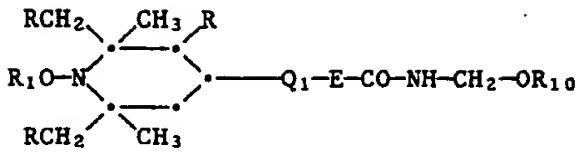
(B)



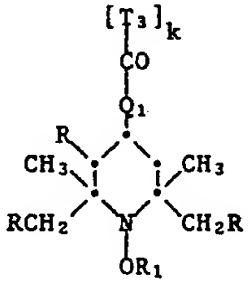
(C)



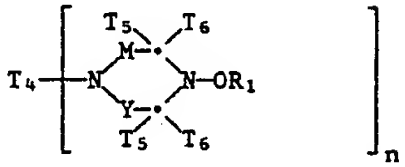
(D)



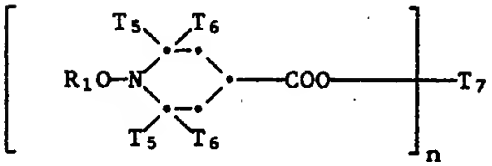
(E)



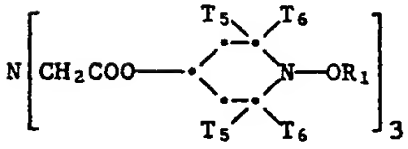
(F)



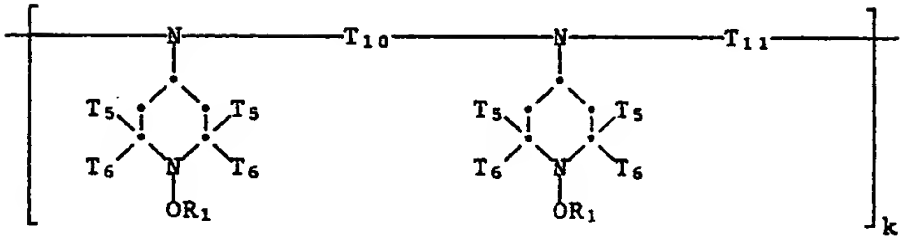
(G)



(H)



(I)



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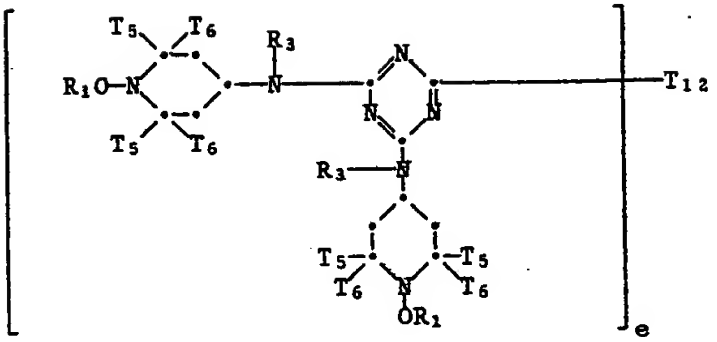
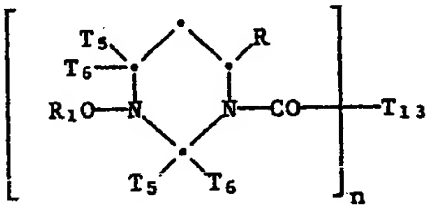


(K)



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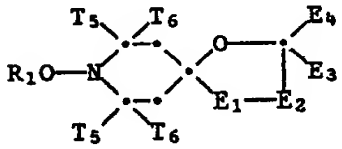

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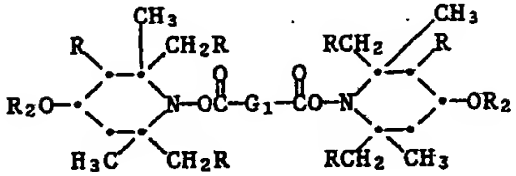
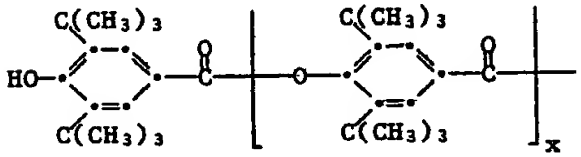
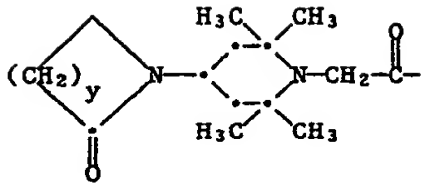


(N)

wherein
R is hydrogen or methyl,
R_1 is a group D-CO- , wherein D is C_1-C_{18} alkyl, C_1-C_{18} alkoxy, phenyl, phenyl substituted by hydroxy, C_1-C_4 alkoxy, or amino or amino mono- or disubstituted by C_1-C_{12} alkyl or phenyl;
m is 1-4,
when m is 1,
R_2 is hydrogen, C_1-C_{18} alkyl optionally interrupted by one or more oxygen atoms, C_2-C_{12} alkenyl, C_6-C_{10} aryl, C_7-C_{18} aralkyl, glycidyl, a monovalent acyl radical of an aliphatic, cycloaliphatic, araliphatic or aromatic carboxylic acid, or of a carbamic acid, or R_2 is a group

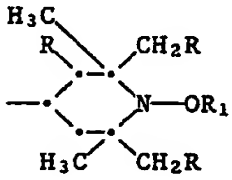


wherein x is 0 or 1, or is a group

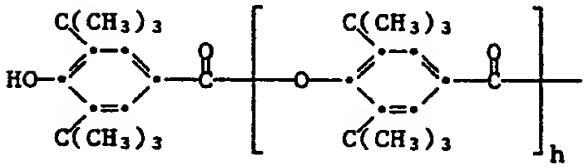


wherein y is 2-4;
when m is 2,
R_2 is C_1-C_{12} alkylene, C_4-C_{12} alkenylene, xylylene, a divalent acyl radical of an aliphatic, cycloaliphatic, araliphatic or aromatic dicarboxylic acid or of a dicarbamic acid, or R_2 is a group

wherein D_1 and D_2 independently are hydrogen, an alkyl radical containing up to 8 carbon atoms, phenyl, benzyl or 3,5-di-t-butyl-4-hydroxybenzyl, and D_3 is an alkyl or alkenyl radical containing up to 18 carbon atoms; when m is 3, R_2 is a trivalent acyl radical of an aliphatic, cycloaliphatic, or aromatic tricarboxylic acid; when m is 4, R_2 is a tetravelent acyl radical of a saturated or unsaturated aliphatic or aromatic tetracarboxylic acid: p is 1, 2 or 3,
R_3 is hydrogen, C_1-C_{12} alkyl, C_5-C_7 cycloalkyl, C_7-C_9 aralkyl, C_2-C_{18} alkanoyl, C_3-C_5 alkenoyl or benzoyl;
when p is 1,
R_4 is hydrogen, C_1-C_{18} alkyl, C_5-C_7 cycloalkyl, C_2-C_8 alkenyl unsubstituted or substituted by a cyano, carbonyl or carbamide group, or it is aryl, aralkyl, glycidyl, a group of the formula $-CH_2-CH(OH)-Z$ or of the formula -CONH-Z wherein Z is hydrogen, methyl or phenyl;
or R_4 is a group of the formula I

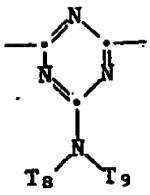


(I) or



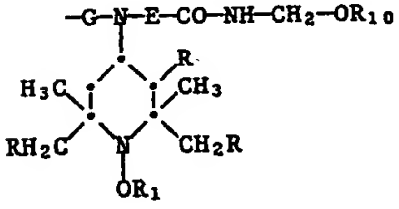
with h as 0 or 1;

or R_3 and R_4 together are alkylene of 4 to 6 carbon atoms or 1-oxo alkylene or the divalent acyl radical of an aliphatic or aromatic 1,2- 1,3-dicarboxylic acid;
when p is 2,
R_4 is C_1-C_{12} alkylene, C_6-C_{12} arylene, xylylene, a $-CH_2CH(OH)-CH_2$ group, or a group $-CH_2-CH(OH)-CH_2-O-X-O-CH_2-CH(OH)-CH_2-$ wherein X is C_2-C_{10} alkylene, C_6-C_{15} arylene or C_6-C_{12} cycloalkylene; or, provided that R_3 is not alkanoyl, alkenoyl or benzoyl, R_4 can also be a divalent acyl radical of an aliphatic, cycloaliphatic or aromatic dicarboxylic or dicarbamic acid, or can be the group -CO-; or R_4 is a group of formula II



(II)

where T_8 and T_9 are independently hydrogen, alkyl of 1 to 18 carbon atoms, or T_8 and T_9 together are alkylene of 4 to 6 carbon atoms or 3-oxapentamethylene, preferably T_8 and T_9 together are 3-oxapentamethylene;
when p is 3, R_4 is 2,4,6-triazinetriyl;
n is 1 or 2 and
when n is 1,
R_5 and R'_5 are independently C_1-C_{12} alkyl, C_2-C_{12} alkenyl, C_7-C_{12} aralkyl, or R_5 is also hydrogen, or R_5 and R'_5 together are C_2-C_8 alkylene or hydroxyalkylene or C_4-C_{22} acyloxyalkylene;
when n is 2,
R_5 and R'_5 together are $(-CH_2)_2C(CH_2-)_2$;
R_6 is hydrogen, C_1-C_{12} alkyl, allyl, benzyl, glycidyl or C_2-C_6 alkoxyalkyl;
when n is 1,
R_7 is hydrogen, C_1-C_{12} alkyl, C_3-C_5 alkenyl, C_7-C_9 aralkyl, C_5-C_7 cycloalkyl, C_2-C_4 hydroxyalkyl, C_2-C_6 alkoxyalkyl, C_6-C_{10} aryl, glycidyl, a group of the formula $-(CH_2)_t$-COO-Q or of the formula $-(CH_2)_t$-O-CO-Q wherein t is 1 or 2, and Q is C_1-C_4 alkyl or phenyl; or
when n is 2,
R_7 is C_2-C_{12} alkylene, C_6-C_{12} arylene, a group $-CH_2CH(OH)-CH_2-O-X-O-CH_2-CH(OH)-CH_2-$ wherein X is C_2-C_{10} alkylene, C_6-C_{15} arylene or C_6-C_{12} cycloalkylene, or a group $-CH_2CH(OZ')CH_2-(OCH_2-CH(OZ')CH_2)_2-$ wherein Z' is hydrogen, C_1-C_{18} alkyl, allyl, benzyl, C_2-C_{12} alkanoyl or benzoyl;
Q_1 is $-N(R_8)-$ or $-O-$;
E is C_1-C_3 alkylene, the group $-CH_2-CH(R_9)-O-$ wherein R_9 is hydrogen, methyl or phenyl, or E is the group $-(CH_2)_3-NH$ or a direct bond;
R_{10} is hydrogen or C_1-C_{18} alkyl;
R_8 is hydrogen, C_1-C_{18} alkyl, C_5-C_7 cycloalkyl, C_7-C_{12} aralkyl, cyanoethyl, C_6-C_{10} aryl, the group $-CH_2-CH(R_9)-OH$ wherein R_9 has the meaning defined above; a group of the formula I
or a group of the formula



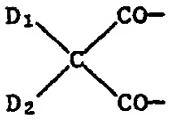
wherein G can be C_2-C_8 alkylene or C_6-C_{12} arylene; or R_8 is a group -E-CO-NH-CH_2-OR_{10};
T_3 is ethylene or 1,2-propylene, or is the repeating structural unit derived from an alpha-olefin copolymer with an alkyl acrylate or methacrylate;
k is 2 to 100;
T_4 has the same meaning as R_4 when p is 1 or 2,
T_5 is methyl,
T_6 is methyl or ethyl, or T_5 and T_6 together are tetramethylene or pentamethylene;
M and Y are independently methylene or carbonyl;
T_7 is the same as R_7;
T_{10} and T_{11} are independently alkylene of 2 to 12 carbon atoms, or T_{11} is a group of formula II;
e is 2, 3 or 4 and
T_{12} is a group $-N(R_5)-(CH_2)_d-N(R_5)-$ or

$-NH(CH_2)_a-\overset{\downarrow}{N}(CH_2)_b-\overset{\downarrow}{N}[(CH_2)_c-\overset{\downarrow}{N}]_fH$ where a, b and c are independently 2 or 3, d is 2 to 10 and f is 0 or 1;
T_{13} is the same as R_4 with the proviso that R_{13} cannot be hydrogen when n is 1;
E_1 and E_2, being different, each are -CO- or $-N(E_5)-$, where E_5 is hydrogen, C_1-C_{12} alkyl or C_4-C_{22}

alkoxycarbonylalkyl;
E_3 is hydrogen, alkyl of 1 to 30 carbon atoms, phenyl, naphthyl, said phenyl or said naphthyl substituted by chlorine or by alkyl of 1 to 4 carbon atoms, or phenylalkyl of 7 to 12 carbon atoms, or said phenylalkyl substituted by alkyl of 1 to 4 carbon atoms,
E_4 is hydrogen, alkyl of 1 to 30 carbon atoms, phenyl, naphthyl or phenylalkyl of 7 to 12 carbon atoms, or E_3 and E_4 together are polymethylene of 4 to 17 carbon atoms, or said polymethylene substituted by up to four alkyl groups of 1 to 4 carbon atoms;
R_2 of formula (N) is as previously defined when m is 1;
G_1 is a direct bond, C_1-C_{12} alkylene, phenylene or -NH-G'-NH wherein G' is C_1-C_{12} alkylene.

3. A composition according to claim 2 which contains a compound of formulae A, B, D, J, K or M wherein R is hydrogen and T_5 and T_6 are methyl.

4. A composition according to claim 2 which contains a compound of formula A wherein R is hydrogen, R_1 is a group D-CO- and D is C_1-C_{12} alkyl, C_1-C_4 alkoxy, phenyl, amino or amino monosubstituted by C_1-C_{12} alkyl or phenyl,
m is 1, 2 or 4 and when m is 1,
R_2 is an acyl radical of an aliphatic C_2-C_{18} carboxylic acid, of a cycloaliphatic C_6-C_{12} carboxylic acid or of an aromatic C_7-C_{15} carboxylic acid and when m is 2, R_2 is a divalent acyl radical of an aliphatic C_2-C_{18} dicarboxylic acid or of a cycloaliphatic or aromatic C_8-C_{14} dicarboxylic acid, or of an aliphatic, cycloaliphatic or aromatic C_8-C_{14} dicarbamic acid, or R_2 is a group



wherein D_1 is C_1-C_8 alkyl or 3,5-di-tert.butyl-4-hydroxybenzyl and D_2 is D_1 or hydrogen and when m is 4, R_2 is a tetravalent acyl radical of a butane- or pentane-tetracarboxylic acid.

5. A composition according to claim 2 which contains a compound of formula B wherein R is hydrogen, R_1 is a group D-CO- and D is C_1-C_{12} alkyl, C_1-C_4 alkoxy, phenyl, amino or amino monosubstituted by C_1-C_{12} alkyl or phenyl,
p is 1 or 2, R_3 is hydrogen, C_1-C_{12} alkyl or C_2-C_{12} alkanoyl and when p is 1, R_4 is C_1-C_{12} alkyl, C_5-C_7 cycloalkyl or a group of formula I,
and when p is 2, R_4 is C_2-C_8 alkylene, phenylene or xylylene, and if R_3 is not alkanoyl, R_4 may also be a divalent acyl residue of an aliphatic C_2-C_{10} dicarboxylic acid or of an aromatic C_6 dicarboxylic acid or of an aliphatic or aromatic C_8-C_{15} dicarbamic acid.

6. A composition of claim 1, which contains di-(1-acetoxy-2,2,6,6-tetramethylpiperidin-4-yl)sebacate.

7. The composition of claim 3, which contains alpha,alpha'-(di-1-ethoxy-2,2,6,6-tetramethylpiperidin-4-yloxy)-p-xylene.

8. The composition of claim 3, containing 1,4-dibenzyloxy-2,2,6,6-tetramethylpiperidine.

9. The composition of claim 3, containing di-(1-benzyloxy-2,2,6,6-tetramethylpiperidin-4-yl) sebacate.

10. The composition of claim 3, containing 4-benzyloxy-1-ethoxy-2,2,6,6-tetramethylpiperidine.

11. The composition of claim 3, containing 1,4-di-(4-hydroxy-3,5-di-tertbutylbenzoyloxy)-2,2,6,6-tetramethylpiperidine.

12. The composition of claim 3, containing alpha,alpha'-(di-1-benzoyloxy-2,2,6,6-tetramethylpiperidin-4-yloxy)-p-xylene.

13. A coating composition according to claim 1, which is an ambient curable system based on an alkyd resin, thermoplastic acrylic resin, acrylic alkyd resin, polyurethane resin or polyester resin, or said resins modified with silicones, isocyanates, epoxides, isocyanurates, ketimines or oxazolidines, or the system is based on a cellulose ester or on an epoxide resin.

14. A coating composition according to claim 1, which is an acid catalyzed thermosetting system based on a hot crosslinkable acrylic, polyester, polyurethane, polyamide or alkyd resin.

15. The composition according to claim 1, which is an enamel for industrial finishes.

16. The composition according to claim 1, which is a refinishing enamel for automobiles.